

## REMARKS/ ARGUMENTS

Reconsideration of the rejection of Specification as failing to provide an adequate written description of the invention and as failing to adequately teach how to make and/or use the invention is respectfully requested, based on the following remarks/arguments.

1. On pages 2,3 Office Action states: "Applicant's invention is at most no more than a variation of the cold fusion concept or system set forth by F and P", where F and P designates Fleischmann and Pons .

Response: Applicant's invention is a variation of the condensed matter nuclear process as set forth by Iwamura et al. (2002) and not as set forth by F and P. The nuclear process set forth by F and P is an electrolytic energized process in which deuterium is loaded in bulk Pd metal cathodes to a abnormally high D/Pd ratio corresponding to a highly non-equilibrium chemical potential. The requirements for a non-equilibrium D/Pd ratio were quantified by McKubre et al. in 1995, [McKubre et al., *Proc. ICCF5*, 17 (1995)] In contrast, the current work of Iwamura et al. does not use electrolysis, and involves relatively low D/Pd ratios. Instead, it involves D/Pd ratios that are in equilibrium with D<sub>2</sub> gas at about 2 atmosphere pressure. The poor reproducibility of studies based on the teaching of F and P is largely due to its non-equilibrium character. This highly non-equilibrium condition causes a deloading instability, which is now understood based on the work of Chernov et al. [I.P. Chernov et al., *Int. J. Hydrogen Energy* **24**, 359 (1999)] and Tyurin and Chernov [Yu.I. Tyurin and I.P. Chernov, *Int. J. Hydrogen Energy* **27**, 829 (2002) ]. These researchers found that metal hydrides can store energy as non-phonon excitations of trapped

hydrogen. The Chernov team showed that such excitation quanta of energy can "diffuse" from an initial disturbance site to distant location on the hydrided metal's surface. They then cause catastrophic hydrogen loss events, with some of the lost hydrogen in atomic rather than molecular form. The hydrogen gas pressure in equilibrium with atomic hydrogen is millions of atmospheres, and can pressurize pores leading to microcracks. The loss of electrolysis-deposited hydrogen as desorbed gas rather than by reversible chemistry is well documented in studies by Farkas [A. Farkas, *Trans. Faraday Society*, **33**, 552 (1937)]. By building on the teachings of Iwamura et al., the Applicant's apparatus and process avoids the highly non-equilibrium chemical potentials and abnormally high D/Pd ratios in bulk Pd that are taught by both F and P and Dash. Applicant argues that his apparatus and process is a variation of F and P only in the sense that both processes result in the radiationless fusion of deuterium into  $^4\text{He}$ .

The teachings of Iwamura et al. differ from those of F and P in other ways. A second difference is that, unlike F and P, Iwamura et al. (2002) do not use heavy water. They use only gaseous deuterium. A third difference is that their proof of nuclear reaction is the observation of a progressing change of surface Cs atoms into surface Pr atoms when deuterium feedstock permeates a Pd reactor plate containing CaO crystallites. The nuclear change is highly exothermic. Control runs have shown that the change does not occur if ordinary hydrogen is used, or if CaO is not present in the reactor plate. A fourth difference is that the Iwamura et al. work was published in a respected peer-reviewed journal, namely the Japanese Journal of Applied Physics [Iwamura et al., *Jpn. J. Appl. Phys.* **41**, 4642 (2002)]. A fifth difference is that the changes in surface composition were observed in a sequence of observations using an in-situ X-ray photoelectron

spectrometer. By using an in-situ spectrometer the reactor plate on which Cs had been deposited was examined by the spectrometer without removal from the reactor containment vessel and without exposure of the surface to air. X-ray photoelectron spectra (XPS) were recorded every few days, measuring the progressive change in surface composition. The in-situ protocol avoids contamination. A lower limit to the nuclear heat generated was calculated from known heats of the reaction sequence effecting  $^{133}\text{Cs} + 8\text{D} \rightarrow ^{141}\text{Pr}$ . The excess heat calculation is valid even though no calorimeter was used. A sixth difference is that a second reaction of the same general type was documented. When surface atoms of Sr were used instead of Cs, there was a reaction sequence effecting  $^{88}\text{Sr} + 8\text{D} \rightarrow ^{96}\text{Mo}$ . A mass spectrometer analysis of post-run Mo recovered from the surface showed that only the mass-96 spectrometer peak had increased.

2. On page 6 Office Action states, "Some members of the scientific community have raised doubts regarding the nuclear process claimed by Iwamura et al. (2002) For example, Shanahan has expressed, among other things, the following items regarding the Iwamura et al.'s claims:

- Reagent purities were not high enough to preclude surface contamination;
- Minimal attempts to eliminate contamination as a source were inadequate;
- Statement that they could detect 0.01 ppm Pr, if present, is an assertion, and the scientific community does not blindly accept a statement without reference."

Applicant argues that Shanahan's contaminant argument is exaggerated and unreasonable. The work was peer reviewed. The in-situ experiment design provided rigorous protection against contamination affecting the XPS measurement process. The 0.01 ppm impurity limit on the bulk materials

making up the reactor plate is an unreasonable requirement considering the surface nature of the detection process. The surface of the Pd reactor plate was initially covered with Cs to about 30% of a monolayer. After the run the Cs had essentially disappeared and there was essentially the same coverage by Pr. There is no chemical process that could have selected a very low level metal impurity from the bulk and selectively carried it to the surface so as to dominate the surface composition. Some have argued that the Pr could have been introduced into the reactor plate in the sputtering process used to create the CaO inclusions. If Pr had been introduced with the CaO, it would have been introduced as  $\text{Pr}_2\text{O}_3$ . There is nothing in the Iwamura process that could have reduced highly stable  $\text{Pr}_2\text{O}_3$  into Pr metal. There is no reasonable explanation of the change from Cs into Pr other than by nuclear reaction. Since the Iwamura et al. (2002) apparatus and process is very different from the cold fusion concept and system set forth by F and P, the decision of the U.S. Federal Court of Appeals *In re. Dash*, No 04-1145, 2004 WL 2829039 (FED. CIR. DEC. 10 2004) is not a valid argument against the Applicant's invention.

3. On page 4 Office Action states: "this 'cold fusion' concept of producing nuclear reactions, including energy generation. (known in the art as 'excess heat'), is still no more than just an unproven concept". On page 9 Office Action states: "....it is incumbent upon the experimenter to show that the alleged experimental results of excess heat and low temperature nuclear products, are valid ....."

Response: The Examiner in the Dash application and the U.S. Federal Court of Appeals failed to adequately consider the knowledge gained in cold fusion studies subsequent to 1994. Subsequent work has demonstrated that there are at

least 3 distinct types of apparatus and process that have provided clearly measured positive values of excess heat in deuterium + palladium experiments.

The first type of apparatus and process is that described by F and P. The F and P studies teach production of excess heat in bulk Pd metal cathodes as a result of over-voltage electrolysis of a D<sub>2</sub>O electrolyte onto a Pd cathode. Palladium deuteride PdD<sub>x</sub> is produced by the electrolysis process. The conditions required for excess heat in this type of equipment were quantified by McKubre et al. in 1995. [McKubre et al., *Proc. ICCF5*, p.17, (1995)] The studies showed that measurable excess heat required an  $x = D/Pd$  ratio  $> 0.83$ , an electrolysis current density  $> 0.4 \text{ A/cm}^2$ , and a time varying D/Pd ratio as measured by  $|dx/dt|$ . The  $|dx/dt|$  term measures the net deuterium flow through the cathode surface, independent of direction. No excess heat was observed during periods when the loading failed to exceed the threshold value and no heat was observed when the loading was not also changing with time. Also, the current density had to exceed  $0.4 \text{ A/cm}^2$ . When all the specified conditions were met, excess heat was observed. In the more successful runs excess heat would turn on and continue for many-hour periods, and then suddenly turn off. In the EPRI M-4 experiment there were two long periods of excess heat, with one lasting 75 hours. The data are shown in an official report requested by DOE [P.L. Hagelstein, M.C.H. McKubre, D.J. Nagel, T.A. Chubb, and R.J. Hekman, "New Physical Effects in Metal Deuterides"], referred to as the "DOE report". A copy of the DOE report was published in *Proc. ICCF11*, p. 23 (2006), subsequent to the filing date. The McKubre empirical excess formula provided a good fit to the two M-4 excess heat periods, as shown in Fig. 5. The M-4 experiment was carried out in a hermetically sealed enclosure, which permitted a search for nuclear product. Fig. 7 is a time history plot of the integral of the excess heat

power, expressed in terms of expected parts per million helium within the hermetic enclosure. It was assumed that the helium production rate would be one helium atom per 23.8 MeV of integrated excess heat energy. The expected helium concentration is compared against 4 mass spectrometer samplings made during the post-run sampling period. Sample 1 measured the helium in the enclosure-contained off-gas shortly after the excess heat periods. Sample 2 was taken about 6 days later, and showed that the helium concentration had slightly grown. Sample 3 was taken about 15 days later and showed the same value as Sample 2. The post-run cathode was then heated and "exercised" to desorb residual helium trapped in the cathode surface region. It was then assumed that all the produced helium had been collected. Sample 4 was then taken. Sample 4 showed that the helium expected from the integrated excess power matched the sum of helium gas in the electrolysis off gas plus the desorbed near-surface gas. The discrepancy fit-error was less than 5%. This good fit between energy release and expected nuclear product seems to be irrefutable evidence for a  $2\text{D} \rightarrow {}^4\text{He}$  radiationless fusion reaction.

Historically, the second type of apparatus and process that has successfully produced cold fusion excess heat is that described by Arata and Zhang (A-Z). Their studies use apparatus and process in which nanometer grains of Pd (nano-Pd) are reacted with deuterium with resulting release of excess heat. In these experiments, a  $\text{PdD}_x$  deuteride is produced by reversible equilibrium chemistry. A-Z have shown that their nano-Pd powders absorb anomalously large amounts of deuterium gas. Some of their powders, when tested for their ability to absorb hydrogen, produced the metal hydride  $\text{PdH}_x$  with values of  $x > 2.9$ , which compares with the maximum non-equilibrium values of  $x < \sim 1.0$  max achieved in bulk metal by the F and P process. A-Z have stated that their deuterium studies

have achieved a 100% success rate in generating excess heat after an initial incubation period.

The A-Z excess heat work of was first published in 1994. [Y. Arata and Y-C Zhang, *Proc. Jpn. Acad.* **70B**, 106 (1994)] The authors used overvoltage electrolysis of a D<sub>2</sub>O electrolyte to deposit deuterons onto the surface of a Pd cylinder containing nano-Pd test powder. The Pd cylinder was closed off at each end to form a pressure tight bottle. The outside of the Pd cylinder served as the electrolysis cell cathode. They called their cell configuration a double structure cathode, designated "DS cathode". The electrolysis-deposited deuterium diffused through the Pd cylinder wall, resulting in a high pressure of D<sub>2</sub> gas in equilibrium contact with the nano-Pd powder. D<sub>2</sub> pressures of over 800 atmosphere were achieved. As shown in their 1994 paper, noticeable heat production began after a 10-day incubation period. It was followed by 12 days averaging above 50 W. The integrated output heat was ~1.8 x integrated input power. In 1995 two additional runs showing continuous excess heat of about 10 W for 100 days were published. (*Proc. Jpn. Acad.* **71B**, 98 (1995)) In 1996 three runs showing ~10 W to 20 W of excess heat were published. [*Proc. ICCF6*, 129 (1996); and *J. High Temperature Society* **23**, 1 (1997)] One of these runs used a pressure gage sampling gas inside the bottle cathode. The gage pinned at 800 atmospheres. In 1998 a pair of DS cathodes were runs in series, with one DS cathode in D<sub>2</sub>O electrolyte and the other in H<sub>2</sub>O electrolyte. The DS cathode in D<sub>2</sub>O electrolyte showed excess heat rising to 20 W, while the one in H<sub>2</sub>O showed no excess heat. [*Jpn. J. Appl. Phys.* **37**, L1274, (1998)]. A-Z prepared a similar pair of DS cathodes containing A-Z processed nano-Pd powder for testing by McKubre and Tanzella at Stanford Research Institute (SRI). The cells were operated by McKubre et al. using the A-Z protocol. Excess heat was measured using SRI calorimetry. The

results obtained by SRI were comparable to those published by A-Z [McKubre et al., *Proc. ICCF8*, 3, (2000), See Fig. 5; and see DOE Report, p. 22]

The continuing series of A-Z excess heat studies produced an especially important run in 2002 . [*Proc. Japan Acad.* **78B**, 57 (2002)] This run made a significant advance by using a new nano-metal material inside their DS cathode. The interior of the Pd cylinder bottle was filled with powder derived from oxidized amorphous  $Zr_{65}Pd_{35}$  alloy. The run was the first to show that powders other than Pd-black can produce excess heat. The new material designated  $ZrO_2$ , nano-Pd, had been manufactured and studied by Yamaura et al. [Yamaura et al., *J. Mater. Res.* 17, 1329 (2002).] This new material was the one that stored hydrogen as  $PdH_x$  with  $x > 2.9$  at 100 atmosphere. X-rays studies showed that the H is not stored as an interstitial in the  $ZrO_2$  lattice. The ground powder is composed of nano-Pd dispersions of 5 to 10 nm dimension inside  $ZrO_2$  crystal. By isolating the nano-Pd crystallites from each other, the Pd crystals cannot grow in size, which causes them to lose their nano-crystalline character. This property is important because of the following: Although the A-Z excess heat process is an equilibrium process, their nano-crystal Pd components are unstable against growth in size. Fig. 4 in the A-Z paper shows steady excess heat generation at 10 W from less than ~ 5 g of Pd. The excess heat production continued steadily for 3.5 weeks, ending at run termination.

Another important A-Z development occurred in 2005. which is subsequent to applicant's filing date. A-Z showed that their  $ZrO_2$ , nano-Pd powder could produce cold fusion heat without the use of electrolysis. A-Z's usual Pd-cylinder bottle was filled with  $ZrO_2$ , nano-Pd powder and was located inside a stainless steel containment vessel which was externally heated to 140 °C. The space



between cylinders was filled with D<sub>2</sub> gas at a pressure of 80 atm. D<sub>2</sub> gas diffused through the Pd cylinder wall and was absorbed by the ZrO<sub>2</sub>, nano-Pd. The resulting cold fusion heat generation raised the temperature of the stainless steel cylinder to a steady state 180 °C, without use of any electrolysis. [*Proc. ICCF12*, ?, (2007)]. Also, the inner Pd cylinder had a higher temperature than the outer stainless steel cylinder, showing that the added heat was being generated in the ZrO<sub>2</sub>, nano-Pd powder, and then flowed outward. The cold fusion heat power has been calculated to be ~ 0.6 W.

The third type of apparatus and process demonstrating radiationless cold fusion energy release is that developed by Iwamura and coworkers. Iwamura et al. (2002) use a difference in D<sub>2</sub> gas pressure to drive a permeation flow of deuterium through a reactor plate with a dispersion of CaO crystallites in 5 internal layers. [Iwamura et al. (2002).] No electrolysis is used. As discussed earlier, their apparatus and process result in a summed exothermic transmutation reaction  $^{133}\text{Cs} + 8 \text{D} \rightarrow ^{141}\text{Pr}$ . The process has been described as occurring in 4 nuclear steps. The first 2 reactions are the same  $2 \text{D} \rightarrow ^4\text{He}$  radiationless fusion reactions demonstrated by McKubre et al. as taught in (T.A. Chubb "Inhibited Diffusion Driven Surface Transmutations")

The Iwamura et al. (2002) permeation studies are an outgrowth of an earlier electrolysis permeation study in which the team seemed to have observed excess heat. [Iwamura et al. (1996)] In a subsequent series of D<sub>2</sub>O-based electrolysis-driven permeation studies using reactor plates containing multiple CaO layers, the Iwamura et al. team observed Watt-level excess heat in 5 experiments. They also observed changes in the surface composition of their reactor cathodes, and

interpreted these new surface atoms as possibly being due to transmutations. [Iwamura et al. (1998)] They then largely switched to a gas pressure-driven permeation process, which enabled them to do in-situ XPS surface analysis on the candidate transmutation products. [Iwamura et al., *Proc. ICCF8*, 141 (2000)] Composition changes were observed, but the changes mostly involved common elements which maybe could have been electrolysis contaminants. This situation made it difficult to prove the transmutation argument. They then decided to use the uncommon element Cs as a starting surface coating, partially because it is an uncommon element. They observed the stepwise change of the Cs to the Pr product, as previously described. Pr is also an uncommon element. [Iwamura et al., *Proc. ICCF9*, 141 (2002)].

4. On page 5 Office Action states: "It was also the general consensus of those skilled in the art and working at these various cold fusion laboratories that there is no reputable evidence of neutron, gamma ray, tritium or helium production to support the allegation or claim that nuclear reactions are taking place, nor is there any reputable evidence to support the allegations or claim of excess heat production."

Response: based on the material presented above it is clear that the work published in 1994 and subsequent years have changed the understanding of the physics underlying cold fusion. The cold fusion field is now understood to be part of the physics discipline Condensed Matter Physics, designated CMP. Cold fusion presentations at the American Physical Society are organized under the Division of condensed matter physics, labeled Dcmp. The international group that conducts the International Conferences of Cold Fusion ( the ICCF meetings) is the International Society for Condensed Matter Nuclear Science (ISCMNS).

Within the broad area of condensed matter physics, the cold fusion art is recognized as being a relative of the physics of electrons in metals.

Those skilled in the Condensed Matter Physics (CMP) art understand that when electrons in an electrochemical cell move from the electrolyte to a metal anode, they undergo a configuration change. Their matter assumes a non-local form. Electron matter in this form is described in the standard solid state textbooks as consisting of "quasiparticles". Electron matter in quasiparticle configuration has no preferred or identifiable center-of-mass. The quanta-of-mass can be said to have a "many-centers-of-mass" form. The quasiparticle mathematical description is called a Bloch function. Those familiar with the CMP art relevant to F and P cold fusion will also know that lab experiments have shown that deuterons on metal surfaces can be configured into the same many-centers form. [M. J. Puska et al., "Quantum Motion of Chemisorbed Hydrogen on Ni Surfaces", *Phys. Rev. Lett.* **51**, 1081, (1983); R. Nieminen, "Hydrogen atoms band together" *Nature* **356**, 289 (1992); C. Astaldi et al., "Vibration Spectra of Atomic H and D on Cu (110): Evidence of H Quantum Delocalization", *Phys. Rev. Lett.* **68**, 909 (1992).]. Those familiar with both the CMP art and the experimental progress described in the DOE report know that energetic neutron and gamma ray emission is not a part of quasiparticle geometry physics. Thus, failure to see neutrons and gamma rays commensurate with excess heat is supportive of F and P type cold fusion. It is not an argument against the F and P process.

There are 2 groups of persons who can be considered skilled in the CMNS art. The first group is those skilled in the CMP art of electrons-in-metals and who also understand the 3 classes of cold fusion experiments described above. The second group is those who lack skills in CMP, yet have both hands-on skills

relevant to laboratory methods discussed thus far, in combination with a knowledge of the experimental results described above plus those discussed in the DOE Report.

Those skilled in the current CMNS art who are familiar with the DOE Report will know that there is reputable evidence for tritium production as a side product in cold fusion processes. Fig. 14 in the DOE Report shows the profile of post-run  $^3\text{He}$  observed from slices of wall material cut from the A-Z cathode operated at SRI. This diffusion-fit profile can only have been created by the decay of tritium as it diffused from the interior of the A-Z cathode to the surrounding electrolyte. Detailed additional evidence is discussed on pages 22 and 23 . The Report states, "The results of gas, metal and electrolyte phase  $^3\text{He}$  and tritium analyses compel the conclusion that tritium was sourced in, on or adjacent to the Pd black in the void of the  $\text{D}_2\text{O}$  double-structure cathode."

Those skilled in the current CMNS art who are familiar with the previously discussed EPRI M-4 experiment will know that there is credible evidence for helium production and commensurate excess heat in F and P type experiments. Those skilled in the current CMNS art who are familiar with the DOE Report will know that there is independent credible evidence for helium production and commensurate excess heat production in diffusion experiments using Case catalyst, as described in DOE Report Appendix B (pp. 18-21). Those skilled in the current CMNS art who are familiar with the previously discussed A-Z studies will know that there is credible evidence for repeatable excess heat. If they are familiar with the larger body of A-Z papers they know that there is credible evidence for associated  $^4\text{He}$  production.

5. Applicant argues non-applicability of pre-1994 art which addresses energetic particle and gamma emission in Judging 2002 art.

Those who judged the credibility of F and P studies prior to 1994 did not have sufficient CMNS skills to judge 2002 CMNS art. They had no basis for understanding that energetic particles and gamma rays are not to be expected in F and P type studies. Those of the pre-1994 References rejecting cold fusion based on failure to observe energetic particle or gamma emission are not relevant to evaluation of the Applicant's invention. The set of non-applicable References cited in the Office Action include the following:

M.H. Salamon et al., "Limits on the emission of neutrons,  $\gamma$ -rays, electrons and protons from Pons/Fleischmann electrolytic cells" , *Nature* **344** (1990).

D. Alber et al., "Search for Neutrons from 'Cold Nuclear Fusion'", *Zeitschrift fur Phys. A, Atomic Nuclei*, **133**, 319 (1989).

P. B. Price et al., "Search for Energetic-Charged-Particle Emission from Deuterated Ti and Pd Foils", *Phys. Rev. Lett.* **63** (1989).

Gad Shani, et al., "Evidence for a Background Neutron Enhancement Fusion in Deuterium Absorbed Palladium", *Solid State Communications* **72**, 53 (1989).

J.F. Ziegler et al., "Electrochemical Experiments in Cold Nuclear Fusion", *Phys. Rev. Lett.* **62**, 2929 (1989).

M. Cribier et al., "Conventional Sources of Fast Neutrons in 'Cold Fusion' Experiments" *Phys. Lett.* **228**, (1989).

S.H. Faller et al., "Investigation of Cold Fusion in Heavy Water, *J. Radioanal. Nucl. Chem . Lett.* **137**, 9 (1989).

J. Hajdas et al., "Search for Cold-Fusion Events", *Solid State Communications*, **72**, 309 (1989) (too low current density)

G. Schrieder et al., "Search for cold nuclear fusion in palladium-deuteride", *Cond. Mat.* **76**, 141 (1989).

G. Chapline, "Cold Confusion", *Proc. NATO Advance Study Institute on the "Nuclear Equation of State"* (1989).

6. Applicant argues non-applicability of pre-1994 art which addresses excess heat production in electrolysis systems that fail to meet requirements specified by McKubre et al.

Prior to post-1994 the experiments that established a quantitative match between integrated excess heat and  $^4\text{He}$  production had not been published. Pre-1994 References cited in the Office Action that operated F and P electrolysis systems at a current density regime or at a D/Pd ratio below that specified by McKubre et al. are not relevant to 2002 CMNS art. Cited References are:

N.S. Lewis et al., "Searches for low-temperature nuclear fusion of deuterium in palladium", *Nature* **340**, (1989). (too low current density)

J. Hajdas et al., "Search for Cold-Fusion Events", *Solid State Communications*, **72**, 309 (1989) (too low current density)

G.M. Miskelly et al., " Analysis of the Published Calorimetric Evidence for Electrochemical Fusion of Deuterium in Palladium", *Science* **246**, 393 or 793 (1989). (too low current density, as per Table 2)

7. Argument that pre-1994 survey and press citations are not relevant to the 2006 art

Prior to 1994 it was believed that nuclear fusion had to be accompanied by energetic particle or gamma emission commensurate with liberated nuclear energy. It was also believed that only the most sensitive and reliable calorimetry had to be used to provide believable excess heat measurements. Post-1994 experiments have established that excess heat could be produced without such emissions and that permeation and nano-metal reactors could produce heat in quantities such that water-flow temperature-rise calorimetry could provide adequate proof of nuclear heat. The pre-1994 survey and press citations express the pre-1994 view and are not relevant to the 2002 CMNS art. Office Action citations in this category not relevant to 2002 art include:

J. F. Cooke, "Report on Foreign Travel", *Letter to A.W. Trivelpiece*, (1989)

M.W. Browne, "'Fusion' Claim is Greeted with Scorn by Physicists", *New York Times*, (1989).

D. Stipp, "Georgia Group Outlines Errors that Led to Withdrawal of 'Cold Fusion' Claims", *Wall Street Journal*, (1989).

P.J. Hilts, "Significant Errors Reported in Utah Fusion Experiments", *The Washington Post*, (1989)

D. Braaaten, "'Ridiculously' easy test yields claim of energy triumph", *Washington Times*, (1989)

Associated Press, "Panel Opposes Cold Fusion Effects", *Washington Post* (1989).

Associated Press, "Physicist: Utah Cold-Fusion Gear Doesn't Work", *Washington Post* (1989).

8. Argument that the permeation reactors of Iwamura et al. and the nano-metal reactors of Arata and Zhang are useful and reproducible, and that their usefulness is not affected by the lack of reproducibility in systems taught by F and P

H. Ohashi and T. Morozumi, "Decoding of Thermal Data in Fleischmann and Pons Paper", *H. Nucl. Sci. and Technolog.* 26, 729 (1989).

Arguments made relative to the lack of reproducibility of the F and P study does not apply to Applicant's invention, which is based on Iwamura et al. (1998 and 2002). Argument that F and P data claiming apparent excess heat can be explained by assuming that hydrogen in the off-gas is oxidized is not likely, because no gas contact with a surface that supports reaction was present in the F and P apparatus. Post-1994 studies establishing match between excess heat and  $^4\text{He}$  increase, used a recombination catalyst and was not affected by oxidation uncertainty. Similarly, the 5 excess heat runs reported by Iwamura et al. using permeation reactors (1998) used a recombination catalyst and was not affected by oxidation uncertainty. Note that the single F and P data point that lies above the Ohashi and Morozumi limit that includes the oxidation hypothesis occurred at a current density above the threshold established by McKubre for F and P



excess heat production. In retrospect, this high current density F and P run may be evidence for excess heat reality.

G. Kreysa et al., "A critical analysis of electrochemical nuclear fusion experiments", *J. Electroanal. Chem.* **266**, 437 (1989).

Argument made relative to the lack of reproducibility of the F and P study does not apply to Applicant's invention, which is based on Iwamura et al. (2002), subsequently found to be closely related to Iwamura et al. (1998). Argument that F and P data claiming apparent excess heat can be explained by assuming that hydrogen in the off-gas is oxidized is not likely, because no gas contact with a surface that supports reaction was present in the F and P apparatus. Post-1994 studies establishing match between excess heat and  $^4\text{He}$  increase, used a recombination catalyst and was not affected by oxidation uncertainty. Similarly, the 5 excess heat runs reported by Iwamura et al. (1998) using permeation reactors used a recombination catalyst and was not affected by oxidation uncertainty.

K.L Shanahan, *Quoted internet conversations with J. Rothwell et al.*, (2002).

Shanahan's questioned claims by Iwamura et al. (2002) arguing that: "Reagent purities were not high enough to preclude surface contamination, minimal attempts to eliminate contamination as a source were inadequate, and statement that could detect 0.01 ppm Pr, if present, is an assertion". Applicant argues that: The surface composition was measured in high vacuum by XPS prior to turn-on of deuterium permeation flow. It was shown to be essentially zero. There are

no known volatile Pr compounds that could have been embedded in the deuterium gas to which the surface was later exposed. The before-start measurement of the surface composition precludes significant Pr surface contamination being initially present, despite Shanahan's arguments. Also, there is no need to establish 0.01 ppm upper limit to Pr in the bulk or near surface volume, or on the surface, because the final observed concentration of Pr exceeds 25% of a mono-layer, which matches the before-start of surface composition of Cs. A selective migration of Pr against such a high concentration gradient is unphysical. Furthermore, if Pr had been present in the initial CaO ionic solid crystallites, it would have been in the form of  $\text{Pr}_2\text{O}_3$ . There is nothing in the Iwamura et al. process that could have chemically reduced it to metal form.

9. Arguments that the revised Specification distinguishes the Applicant's invention from apparatus References cited in the Office Action of 24 November 2006.

E. Granite and J. Jorne, "A novel method for studying electrochemically induced cold fusion using a deuteron-conducting solid electrolyte", *J. Electroanal. Chem.* 317, 285 (1991).

The Reference describes use of a  $\beta$ -alumina solid electrolyte option that could be used in the Applicant's invention as input electrolysis and output electrolysis cells in Applicant's apparatus. However, the teachings of Granite and Jorne do not include the essential reactor plate containing ionic solid inclusions specified in the Applicant's apparatus. The Granite and Jorne lacks an essential ingredient taught in the Applicant's invention, and cannot support the Applicant's process.

A. V. Joshi. "ELECTROLYTIC APPARATUS FOR DISASSOCIATION OF COMPOUNDS CONTAINING HYDROGEN ISOTOPES", *U.S. Patent No. 4,725,346* (1988).

The Reference describes use of solid electrolyte cells that could be used in the Applicant's invention as input electrolysis and output electrolysis cells in Applicant's apparatus. However, the teachings of Joshi do not include the essential permeation reactor plate containing ionic solid inclusions specified in the Chubb apparatus. The Joshi apparatus by itself cannot support the Applicant's process. The Joshi apparatus as claimed requires use an anode pervious to oxygen, which is avoided in Applicant's invention.

F. David, "DEVICE FOR CONTAINING NUCLEI OF LIGHT ATOMS IN A SOLID PHASE", *French Patent No. 2.662,537 A1*, (1994).

F. David teaches an apparatus that specifies an electrode reactor that contains silicon or other p-type semiconductor contacting Pd or other metal. Applicant teaches an apparatus that specifies a reactor containing ionic solids contacting Pd or other metal. The semiconductor-metal contact used by David cannot function in the Applicant's invention. The modeling of the David process cannot explain the Iwamura et. al. (2002) production of nuclear reactions, because the David process teaches production of energetic particles commensurate to nuclear energy release, whereas the Iwamura et al. (2002) process does produce energetic particles.

Sugaya et al., "Gas sensor", *U.S. Patent No. 6,635,162 B2*.

Sugaya et al. teach a process involving flowing HD combustible gas. Applicant's invention avoids any gas flow that includes a hydrogen-carbon compound. Sugaya et al. do not use an interior electrode or reactor plate that contains ionic solid crystals like those composed of CaO. Ionic solid crystallites fully embedded inside a metal are essential parts of the apparatus specified in the Applicant's invention. Sugaya et al. use diffusion controlling passages that provide means by which gas being tested moves by diffusion to an internal gas-filled volume in contact with solid electrolyte cells. The diffusion process referred to is gas molecular diffusion through small passages and pores in a body made by firing a paste made of Alumina powder, an organic binder and an organic solvent. There is no metal in the paste. The diffusion process employed in Applicant's invention is diffusion of hydrogen ions within a metal crystal, and does not involve pores. The word "diffusion" means flow induced by a gradient of a particle density distribution. The particle density distribution in Sugaya et al. refers to molecules of a gas. The particle density in the Application refers to D<sup>+</sup> ions embedded inside a metal in interstitial trapping sites where conduction electrons of the metal neutralize the trapped ion charge. This definition is recognized in the hydrogen-in-metals art.

The Sugaya et al. porous insulator described in the Office Action cannot be read as crystals dispersed in a metal, because the porous insulator lacks the crystal-metal interfaces that provide the catalytic environment that causes the deuteron to change its configuration to a nuclear active form. Also, pores are not part of the Applicant's invention. In the Applicant's invention the word "scatterings" refers only to interactions with ionic crystal-metal interfaces.

#### References used in Remarks Concerning Specification

Y. Arata and Y.-C. Zhang, "A New Energy caused by 'Spillover-Deuterium'", Proc. Japan Acad. 70B, p. 106 (1994); Y. Arata and Y.-C. Zhang, "Solid State Plasma Fusion ('Cold Fusion')", *J. High Temperature Soc. Japan.* 23 (Special Vol.), p. 1 (1998).

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T. A. Chubb. Copy of notes written by T. Chubb during Iwamura talk at ICCF11 in Marseilles, France, 2004. Enclosed.

A. De Ninno, A. Frattolillo, Z Rizzo, E.. Del Giudice, and G. Preparata, "Experimental Evidence of the  $^4\text{He}$  production in a Cold Fusion Experiment", (ENEA Centro Ricerche Frascati. C.P. 65 - 00044 Frascati, Rome, 2002)

M. McKubre, F. Tanzella, P. Tripodi and P. Hagelstein, "The Emergence of a Coherent Explanation for Anomalies Observed in D/Pd and H/Pd Systems: Evidence for  $^4\text{He}$  and  $^3\text{H}$  Production", Proc. ICCF8, F. Scaramuzzi Editor (Italian Physical Society, Bologna, 2000) p. 3.

M. H. Miles and B. F. Bush, "Heat and Helium Measurements in Deuterated Palladium", *Trans. Fusion Technol.*, 26, p. 156 (1994).

Puska, M. J., Nieminen, J. R. M., Manninen, M., Chakraborty, B., Holloway, S. & Norskov J. K. 1983 Quantum Motion of Chemisorbed Hydrogen on Ni Surfaces. *Phys. Rev. Lett.* 51, 1081-1084.

Puska, M. J. & Nieminen, R. M. 1985 Hydrogen chemisorbed on nickel surfaces: a wave mechanical treatment of proton motion. *Surface Science* **157**, 413-435.

#### Arguments against Claim Rejections

Reconsideration of the rejection of Claims 9 - 12 is respectfully requested.

#### *Claim Rejections - 35 USC §101*

Reconsideration of the rejection of Claim 1 under 35 USC 101 is respectfully requested because the Applicant's invention uses permeation plate reactor technology demonstrated by Iwamura et al. (1998), which technology demonstrated excess heat in 5 runs. Also the Applicant's invention shares gas purity and permeation technology with Iwamura et al. (2002), which demonstrated exothermic nuclear reactions and which was published in the peer-reviewed Japan Journal of Applied Physics. The invention's electrolysis deuterium permeation drive can use a solid-electrolyte electrolysis cell of the type described by Biberian (2002), which used accepted fuel cell solid-electrolyte technology. The Biberian cell functioned in the manner expected in accord with electrochemical theory. But Biberian did not teach use of electrolysis to power permeation flow. The use of solid-electrolyte electrolysis to power permeation flow through a reactor plate containing ionic solid inclusions is specified in Applicant's invention.

At a minimum, the Applicant's invention has utility as a test bed to enable reliable comparison of the heat generation capabilities of test permeation plate reactors that differ in design detail, such as in their internal distribution of ionic solid-metal interfaces and the compositions, shapes and area of the interfaces provided. The invention is therefore useful as a development tool for deuterium

fusion heaters. The invention improves on the technology used by Iwamura et al. (1998) in that it increases excess power by reducing parasitic electric power, while maintaining gas purity comparable to that used in Iwamura et al. (2002). It improves on the technology used by Iwamura et al. (2002) by supporting operation at higher deuterium chemical potential and higher permeation flow rate. It is likely that the Applicant's invention will have utility as a commercial cold fusion heater once lower cost materials and fabrication techniques are identified.

The argument made against the Applicant's invention that "the invention is based upon allegations that border on the incredible" are outdated. About half of the DOE Review Panel tasked with judging the DOE Report concluded that the evidence for excess heat was valid. They recommended continued research. The Panel's split acceptance of the excess heat evidence occurred despite the fact the almost none of the reviewers were familiar with cold fusion art as it has developed since 1994. Also, none of the Iwamura et al. work was presented for review, and was included in the DOE Report only as a Reference. Also, Arata and Zhang work subsequent the DOE Report shows that heat production can occur in deuterided nano-Pd without use of electrolysis, i.e., at relatively low deuterium chemical potential.

### *Claim Rejections - 35 USC §112*

Reconsideration of the rejection of Claim 9-12 under 35 USC 112 is respectfully requested because the Specification as now revised is believed to adequately describe the subject matter. Previous deficiencies have been addressed in such a way as to enable one skilled in the art to make and use the invention. The revised specification adequately specifies the need to have a permeation reactor plate that contains a dispersion of ionic solid-metal interfaces.

The periodic order provided by the ionic solid at one side of the interface is an essential feature of the invention. The negative charge provided by the mobile electrons of the metal that neutralize the interface deuterium ions at the second side of the interface volume is an essential feature of the invention. These two ingredients are enabling, when combined with the apparatus and process described in the Specification.

The revised specification adequately specifies the invention's avoidance of liquid electrolytes, and the avoidance of electrolytes containing deuterium oxides. Previously used D<sub>2</sub>O-based systems require higher electrolysis cell voltages, because they split D<sub>2</sub>O into D<sub>2</sub> + O<sub>2</sub>, which increases parasitic power loss. Liquid-electrolyte electrolysis also increases the likelihood of deposition of impurities on the inflow surface of the permeation reactor plate. Applicant's invention teaches closed-loop operation as an essential feature because it reduces contamination possibilities and keeps circulation power low.

#### ***Claim Rejections - under 35 USC §102***

Reconsideration of the rejection of Claim 9 under 35 USC 102 as anticipated by Joshi is respectfully requested. Joshi describes use of solid electrolyte cells that could be used in the Applicant's invention as input electrolysis and output electrolysis cells in Applicant's apparatus. However, the teachings of Joshi do not include a permeation reactor plate containing ionic solid inclusions as specified in the invention apparatus. The invention-specified permeation reactor plate is an essential component of the invention. Joshi's use of carbon fibers does not apply because the carbon fibers are not ionic solids and do not impose the same alternating charge polarity potential leads to order in the ionic solid crystallite-metal interface. The Joshi apparatus by itself cannot support the Applicant's process. Also, the Joshi apparatus as claimed requires



use of an anode pervious to oxygen. The Applicant's invention specifically avoids use of any oxidized deuterium compound and avoids use of an anode pervious to oxygen. When an oxidized deuterium compound is employed, oxygen is produced at the anode. When all oxidized deuterium compounds are avoided, there is no electrolytic oxygen produced. In contrast to the Applicant's process, the Joshi system does not reduce the parasitic electric power required to electrolyze  $D_2O$  in  $LiOD$  or  $LiOH$ , and does not avoid deposition of contaminants dissolved in liquid electrolytes. The Applicant's invention eliminates the parasitic power required to electrolyze  $D_2O$  and avoids deposition of contaminants dissolved in any liquid electrolyte.

Reconsideration of the rejection of Claims 9 - 12 under 35 USC 102 as anticipated by Sugaya et al. is respectfully requested. Sugaya et al. teach a process involving flowing HD combustible gas molecules. Applicant's invention avoids any gas flow that includes a hydrogen-carbon compound. Sugaya et al. do not use an interior electrode or reactor plate that contains ionic solid crystals like those composed of  $CaO$ . Ionic solid crystallites inside a metal are essential parts of the apparatus specified in the Applicant's invention. Sugaya et al. use diffusion controlling passages that provide means by which gas being tested moves by diffusion to an internal gas-filled volume in contact with solid electrolyte cells. The diffusion process referred to by Sugaya et al. is gas molecular diffusion through small passages and pores in a ceramic body made by firing a paste made of Alumina powder, an organic binder and an organic solvent. There is no metal in the paste. The diffusion process employed in Applicant's invention is diffusion of interstitial hydrogen ions within a metal crystal. Interstitial ion diffusion in metals is fundamentally different from gas molecule diffusion through porous ceramic. The diffusion in the Applicant's invention does not involve pores. The Sugaya et al. porous insulator described

in the Office Action cannot be read as crystals dispersed in a metal, because the porous insulator lacks the ionic solid crystallite-metal interfaces that provide the catalytic environment that causes the deuteron to change its configuration to a nuclear active form. Pores are not part of the Applicant's invention. In the Applicant's invention scatterings refer to configuration-changing non-pore interactions with ionic crystal-metal interfaces. Sugaya et al. do not teach the required use of a metal plate reactor containing salt-metal interface volumes such as are present when the reactor contains a dispersion of ionic solids inside a metal plate.

#### *Claim Rejections - 35 USC §103(a)*

Reconsideration of the rejection of Claim 10 under 35 USC 103a as unpatentable over Joshi in view of McIntyre et al. (U.S. 4,279,709) is respectfully requested. The McIntyre et al. invention is titled "Preparation of Porous Electrodes". It teaches the use of soluble bodies applied as temporary components of coating applied to electrolysis cell electrodes. One of the solids suggested as a coating component is CaO. Although CaO is a preferred ionic solid suggested for use in Applicant's invention, there is no relation between the Applicant's use of CaO and its use in McIntyre et al., because the CaO as used in Applicant's invention specifies that it be embedded in metal, whereas its use in McIntyre et al. requires that it be exposed to a surface and be subsequently dissolved in an electrolyte to create a pore. It is embedded in a permeation plate reactor and makes no contact with the surface of the reactor plate, and no contact with any electrolyte. Also, there is no porous electrode used in the Applicant's patent. Adding the teaching of McIntyre et al. to the teaching of Joshi does not enable Joshi to read on the Applicant's invention, because neither teach use of a permeation reactor plate containing ionic solid inclusions.

Reconsideration of the rejection of Claim 12 under 35 USC 103a as unpatentable over Joshi in view of Idota et al. (U.S. 4,279,709) is respectfully requested. The Idota et al. invention is titled "Nonaqueous Secondary Battery Containing Silicic Material". It teaches the use of silicic material as a component of the negative electrode in a Li battery. The silicic material stores Li when the battery is charged. They teach the use of such a negative electrode in conjunction with a solid electrolyte containing polypropylene oxide polymer and phosphoric acid ester polymer mixture. The use of this teaching in the Joshi invention does not affect the of the Joshi invention on the Applicant's invention because the Joshi invention is not readable on the Applicant's invention because the teachings of Joshi do not include a permeation reactor plate containing ionic solid inclusions as specified in the Applicant's apparatus.

For the above reasons, applicant believes that the amended specification and claims now set forth the invention adequately and that the claims are patentable over the prior art.

Respectfully Submitted,

A handwritten signature in cursive script that reads "Melvin L. Crane". The signature is written in dark ink and is positioned above the printed name and registration number.

Melvin L. Crane  
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DOE

December 1, 2004

## Report of the Review of Low Energy Nuclear Reactions

### Introduction

The Department of Energy's (DOE) Office of Science (SC) was approached in late 2003 by a group of scientists who requested that the Department revisit the question of scientific evidence for low energy nuclear reactions. In 1987 Pons and Fleischman first reported the production of "excess" heat in a Pd electrochemical cell, and postulated that this was due to D-D fusion (D=deuterium), sometimes referred to as "cold fusion." The work was reviewed in 1989 by the Energy Research Advisory Board (ERAB) of the DOE. ERAB did not recommend the establishment of special programs within DOE devoted to the science of low energy fusion, but supported funding of peer-reviewed experiments for further investigations. Since 1989, research programs in cold fusion have been supported by various universities, private industry, and government agencies in several countries.

### Review and Process

In response to the above request, the Office of Science agreed to a peer review of the experimental data and supporting theory since the 1989 ERAB review. The scientists who made this request were asked to generate a review document that identified the most significant experimental observations and publications, and those areas where additional work would appear to be warranted. This document, entitled "New Physical Effects in Metal Deuterides," was prepared by Professor Peter Hagelstein of MIT, Dr. Michael McKubre of SRI International, Professor David Nagel of George Washington University, Dr. Talbot Chubb of Research Systems Inc., and Mr. Randall Hekman of Hekman Industries (hereafter referred to as the proposers). Together with supplemental material, said document was submitted to DOE in July 2004 (Attachment 1).

The Basic Energy Sciences and Nuclear Physics Offices in the DOE Office of Science conducted a peer review of the submitted material in a manner typical for a DOE sponsored university or laboratory research program. The review had two components. First, the review document received by DOE was sent out for peer review by mail. Nine scientists with appropriate scientific backgrounds in experimental and theoretical nuclear physics, material science, and electrochemistry were identified by DOE, and were given approximately one month to review the report and supplementary material. The second part of the review consisted of a one-day review conducted on August 23, 2004. The reviewers consisted of nine additional scientists chosen by DOE for their expertise in relevant fields. Anonymous comments from the mail peer review referred to above were provided to members of the reviewers prior to the presentations. Oral presentations were made to the reviewers by research scientists, chosen by the authors of the review document. Six research groups gave approximately one hour presentations on the work being performed in their laboratories. Individual comments from reviewers were requested following the presentations.

In total, 18 individual reviewer comments were received by DOE.

### Review Criteria

Reviewers were asked to respond to the following charge in their evaluation of the written and/or oral material: (1) To examine and evaluate the experimental and theoretical evidence for the occurrences of nuclear reactions in condensed matter at low energies (less than a few electron volts). (2) To determine whether the evidence is sufficiently conclusive to demonstrate that such nuclear reactions occur. (3) To

determine whether there is a scientific case for continued efforts in these studies and, if so, to identify the most promising areas to be pursued. Copies of the charge letter and accompanying instructions regarding conflict of interest and confidentiality are attached (Attachment 2).

### **Review Document and Presentations**

The review document submitted (Attachment 1) focused on "a subset of research from two areas" in the field of low energy nuclear reactions: (1) "selected issues associated with excess heat production in deuterated metals" and (2) "some aspects of nuclear emissions from deuterated metals." According to the review document, D-D fusion has been demonstrated to occur spontaneously when D is introduced into Pd metal at very high concentrations ( $D/Pd \sim 0.95$ ). According to the review document, these demonstrations include purported production of anomalous energy, helium, tritium, and a variety of elements not initially present in the experimental container.

The material presented in the review document and oral presentations focused on electrochemical reactions in the Pd/D<sub>2</sub>O system, evidence for excess heat and nuclear reaction products, and the current theoretical framework that has been used to describe the observations. Data were also presented on the use of ion beams and glow discharge systems used to study the Pd,Ti/D and Pd,Ti/H systems. The review only addressed "light element" experiments, namely H or D fusion.

The proposers state that the results from the research provide evidence for effects in three categories, as summarized in the review document's Conclusions Chapter:

1. "The existence of a physical effect that produces heat in metal deuterides. The heat is measured in quantities greatly exceeding all known chemical processes and the results are many times in excess of determined errors using several kinds of apparatus. In addition, the observations have been reproduced, can be reproduced at will when the proper conditions are reproduced, and show the same patterns of behavior. Further, many of the reasons for failure to reproduce the heat effect have been discovered."
2. "The production of <sup>4</sup>He as an ash associated with this excess heat, in amounts commensurate with a reaction mechanism consistent with  $D+D \rightarrow {}^4\text{He} + 23.8 \text{ MeV (heat)}$ ".
3. "A physical effect that results in the emission of: (a) energetic particles consistent with  $d(d,n){}^3\text{He}$  and  $d(d,p)t$  fusions reactions, and (b) energetic alphas and protons with energies in excess of 10 MeV, and other emissions not consistent with deuteron-deuteron reactions."

The material presented can be found at <http://www.sc.doe.gov>. Following the oral presentations, reviewers requested additional documentation from the presenters. This supplemental material can also be found at the indicated link.

### **Detailed Summary of Reviewer Response to Charge Elements**

Since the 1987 report by Pons and Fleishman, scientists have continued to investigate the conditions responsible for the anomalous heat production in an attempt to establish reproducible conditions for the generation of excess energy, quantify the amount of energy being released, and confirm the hypothesis that the energy is a consequence of nuclear fusion by detecting the expected nuclear reaction products. Below is a summary of the reviewer responses to the three charge elements, written by DOE program managers and intended to give an overall sense of the reviewers' comments. The entire charge letter is

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enclosed as Attachment 2. The redacted reviewer comments (only their names and institutions were removed) have been sent to the proposers.

**Charge Element 1: Examine and evaluate the experimental evidence for the occurrences of nuclear reactions in condensed matter at low energies (less than a few electron volts).**

The experimental evidence presented by the review document and oral presentations for the occurrences of nuclear reactions consisted of two general types: excess power production from an electrolytic cell containing metal electrodes (palladium is the typical metal) with a deuterated electrolyte; and measurement of expected fusion products such as  $^4\text{He}$  in electrolytic cells, or any of the other expected products observed in hot fusion, proton + triton (the nucleus of tritium, consisting of two neutrons and one proton) or neutron +  $^3\text{He}$ , in a variety of experiments.

The excess power observed in some experiments is reported to be beyond that attributable to ordinary chemical or solid state sources; this excess power is attributed by proponents to nuclear fusion reactions. Evaluations by the reviewers ranged from: 1) evidence for excess power is compelling, to 2) there is no convincing evidence that excess power is produced when integrated over the life of an experiment. The reviewers were split approximately evenly on this topic. Those reviewers who accepted the production of excess power typically suggest that the effect seen often, and under some understood conditions, is compelling. The reviewers who did not find the production of excess power convincing cite a number of issues including: excess power in the short term is not the same as net energy production over the entire of time of an experiment; all possible chemical and solid state causes of excess heat have not been investigated and eliminated as an explanation; and production of power over a period of time is a few percent of the external power applied and hence calibration and systematic effects could account for the purported net effect. Most reviewers, including those who accepted the evidence and those who did not, stated that the effects are not repeatable, the magnitude of the effect has not increased in over a decade of work, and that many of the reported experiments were not well documented.

The hypothesis that excess energy production in electrolytic cells is due to low energy nuclear reactions was tested in some experiments by looking for  $\text{D} + \text{D}$  fusion reaction products, in particular  $^4\text{He}$ , normally produced in about 1 in  $10^7$  in hot  $\text{D} + \text{D}$  fusion reactions. Results reported in the review document purported to show that  $^4\text{He}$  was detected in five out of sixteen cases where electrolytic cells were reported to be producing excess heat. The detected  $^4\text{He}$  was typically very close to, but reportedly above background levels. This evidence was taken as convincing or somewhat convincing by some reviewers; for others the lack of consistency was an indication that the overall hypothesis was not justified. Contamination of apparatus or samples by air containing  $^4\text{He}$  was cited as one possible cause for false positive results in some measurements.

Beam experiments not involving electrolytic cells were reported in the review document and oral presentation, purport to provide evidence for low energy nuclear reactions. These experiments involved low energy deuterium beams impinging on deuterium loaded metal foils such as titanium. The studies were designed to investigate screening effects in materials that would be relevant to fields such as nuclear astrophysics. Those reviewers who commented on these studies generally viewed them favorably, but to many reviewers these studies were somewhat peripheral to the main thrust of this review.

A similar line of investigation involved counting deuterium loaded foils to observe the products for the standard fusion reaction channels, proton + triton or neutron +  $^3\text{He}$ , with particle detectors and coincidence techniques. Indications of purported detection of proton-triton coincidences at a low level were presented. Even skeptical reviewers cited this work as one line of investigation that could be pursued to a clear conclusion. However, the results were not convincing to some reviewers in regard to the occurrence of low energy nuclear reactions. Experts noted many deficiencies in the techniques, methods, and interpretation of the data presented. The present state-of-the-art for tracking coincidences and the methodology for low data rate experiments is far advanced beyond methods used in the experiment contained in the review document and oral presentations.

Two-thirds of the reviewers commenting on Charge Element 1 did not feel the evidence was conclusive for low energy nuclear reactions, one found the evidence convincing, and the remainder indicated they were somewhat convinced. Many reviewers noted that poor experiment design, documentation, background control and other similar issues hampered the understanding and interpretation of the results presented.

**Charge Element 2: Determine whether the evidence is sufficiently conclusive to demonstrate that such nuclear reactions occur.**

Reviewers expert in nuclear physics noted that the cold fusion mechanism put forward by proponents is not in accord with presently accepted knowledge of D + D fusion. Specifically, D + D fusion is accompanied by the production of protons, neutrons, tritons,  $^3\text{He}$ ,  $^4\text{He}$  and high energy gamma rays, all in well known proportions. The fusion channel resulting in  $^4\text{He}$  and high energy gamma rays occurs approximately only once for every  $10^7$  D + D fusion reactions. These characteristic proportions for the production of the fusion products are found for every energy of the incident deuteron measured so far, down to the lowest that has been measured.

The review document and oral presentations made the argument that the branching ratios are different at low energies and that in cold fusion,  $^4\text{He}$  fusion channel is predominant. According to the review document, no high energy gamma rays appear to accompany the  $^4\text{He}$ , as is observed in D-D fusion reactions. Instead, the approximately 24 MeV in energy resulting from D-D fusion was purported to appear as heat in the material lattice. To explain these unusual characteristics, the reviewers were presented with a theoretical framework that purported to describe how collective energy from the material lattice couples to a deuteron pair to induce fusion, how the only fusion reaction channel that occurs would be the production of  $^4\text{He}$ , and how all the energy is coupled back into the material in the form of heat instead of high energy gamma-rays. The reviewers raised serious concerns regarding the assumptions postulated in the proposed theoretical model for the explanation for  $^4\text{He}$  production.

The preponderance of the reviewers' evaluations indicated that Charge Element 2, the occurrence of low energy nuclear reactions, is not conclusively demonstrated by the evidence presented. One reviewer believed that the occurrence was demonstrated, and several reviewers did not address the question.

**Charge Element 3: Determine whether there is a scientific case for continued efforts in these studies and, if so, to identify the most promising areas to be pursued.**



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The nearly unanimous opinion of the reviewers was that funding agencies should entertain individual, well-designed proposals for experiments that address specific scientific issues relevant to the question of whether or not there is anomalous energy production in Pd/D systems, or whether or not D-D fusion reactions occur at energies on the order of a few eV. These proposals should meet accepted scientific standards, and undergo the rigors of peer review. No reviewer recommended a focused federally funded program for low energy nuclear reactions.

Reviewers identified two areas where additional research could address specific issues. One is the investigation of the properties of deuterated metals including possible effects of alloying and dislocations. These studies should take advantage of the modern tools for material characterization. A second area of investigation is the use of state-of-the-art apparatus and techniques to search for fusion events in thin deuterated foils. Several reviewers specifically stated that more experiments similar in nature to those that have been carried out for the past fifteen years are unlikely to advance knowledge in this area.

## **Conclusion**

While significant progress has been made in the sophistication of calorimeters since the review of this subject in 1989, the conclusions reached by the reviewers today are similar to those found in the 1989 review.

The current reviewers identified a number of basic science research areas that could be helpful in resolving some of the controversies in the field, two of which were: 1) material science aspects of deuterated metals using modern characterization techniques, and 2) the study of particles reportedly emitted from deuterated foils using state-of-the-art apparatus and methods. The reviewers believed that this field would benefit from the peer-review processes associated with proposal submission to agencies and paper submission to archival journals.

Attachment 1: Review document submitted by requesters, "New Physical Effects in Metal Deuterides."  
Attachment 2: Charge letter to reviewers

## Nuclear Physics Approach

### **Correlation between Behavior of Deuterium in Palladium and Occurrence of Nuclear Reactions Observed by Simultaneous Measurement of Excess Heat and Nuclear Products**

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#### Abstract

We developed a new type of experimental apparatus for simultaneous measurement of excess heat and nuclear products with intent to induce continuous nuclear reactions in D<sub>2</sub>-Pd system. It consists of two parts; an electrochemical cell for calorimetry and a vacuum chamber for nuclear measurement. There exists continuous flow of deuterium atoms from the electrochemical side to the vacuum side through a palladium plate and we might expect that nuclear reactions last for a long term in the system, since it is considered that diffusion process of deuterium atoms in palladium is important to induce nuclear reactions.

Our experimental results suggest that absorption and desorption behavior of deuterium was greatly influenced by unspecified factors (impurity or metallurgical conditions of palladium). X-ray emission lasting for long term and excess heat generation was observed, however, correlation between excess heat and x-ray is not clear under our experimental condition.

#### 1. Introduction

We previously reported that diffusion process of deuterium, in addition to high D/Pd ratio, is important factor for causing nuclear reactions by the method of gas release experiments in vacuum<sup>1,2</sup>. Based on these experimental results, we developed a new experimental apparatus for simultaneous measurement of excess heat and nuclear products.

The basic idea of our apparatus is that we might make continuous nuclear reactions occur in D<sub>2</sub>-Pd system by making continuous flow of deuterium under *certain appropriate conditions*. Although high D/Pd ratio is one of the *certain appropriate conditions*, the other necessary conditions still remain to be unclarified at present. Therefore it might be possible to clarify the other *appropriate conditions* by making continuous flow of deuterium in palladium under high D/Pd condition; For it is considered to be most probable way to induce nuclear reactions according to our experimental results<sup>1-3</sup>.

In this paper, we describe our experimental apparatus and the results on simultaneous measurement of excess heat and nuclear products.

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## 2. Experimental Method

Fig.1 shows a cross sectional view of the experimental apparatus. An electrolyte of 1M LiOD/D<sub>2</sub>O and a vacuum chamber are separated by a palladium plate. Deuterium atoms are loaded by electrochemical potential into one side of the palladium sample and released from the other side.

Excess heat is estimated by the flow calorimetry method. Electrolyte side of the apparatus consists of a cathode of palladium plate (25x25x1mm; 99.9% Tanaka Kikinzoku Kogyo K.K.), a circular shape of anode of platinum mesh (φ1mm), a recombiner and a cooling pipe for measurement of excess heat generation. Two thermocouples each for measurement of inlet and outlet temperature of the water are provided. Solution, gas, recombiner and environmental temperatures are measured and consistency between these temperatures is always checked.

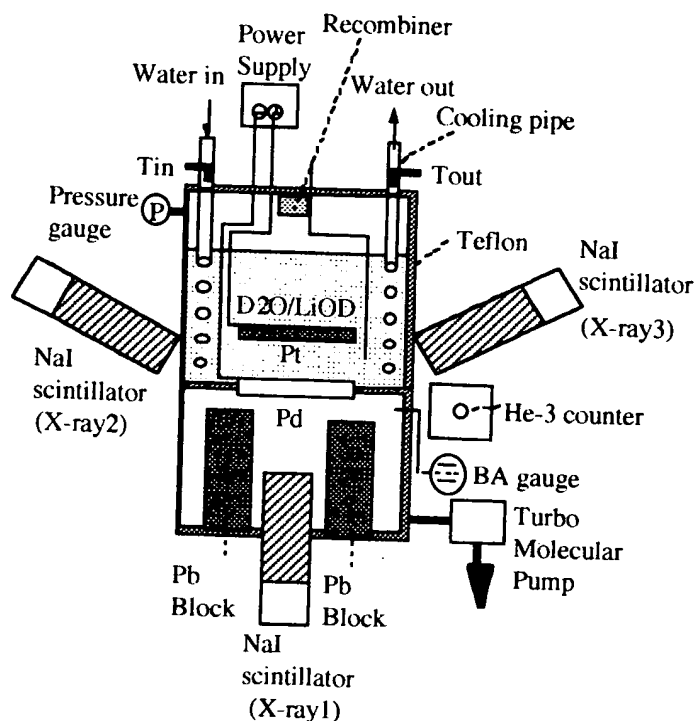


Fig.1 Experimental Apparatus

The apparatus is equipped with three NaI scintillation counters (Bicron: 1.5XM1/2B) for x-ray spectroscopy and a He-3 neutron detector (Reuter-Stokes: RS-0806-207). The X-ray1 detector located in the vacuum chamber is surrounded by a lead cylinder (thickness; 2.5cm) to reduce background x-rays. X-ray spectroscopy and counting system consists of preamplifiers (EG&G Ortec : 276), amplifier and single channel analyzers (EG&G Ortec : 590A), counters (EG&G Ortec : 997) and multi-channel analyzers (SEIKO EG&G : MCA4100, 4200). As to neutron counting, we use a preamplifier (EG&G Ortec : 142PC), an amplifier and single channel analyzer (EG&G Ortec : 590A) and a counter (EG&G Ortec : 997).

Pressures of the upper part of electrolyte and vacuum chamber are monitored by a pressure gauge and a BA gauge, respectively. D/Pd is calculated by the obtained pressures as will be described. All the experimental data is acquired by personal computers.

## Nuclear Physics Approach

The apparatus and measuring systems are located in a clean-room where temperature and humidity are always controlled at constant levels ( $23^{\circ}\text{C} \pm 1^{\circ}\text{C}$ ,  $40\% \pm 5\%$ ). Furthermore, all these electric devices are supplied by an isolated power source to prevent electric noise from the outside of the clean room.

Procedure of sample preparation is as follows. Palladium plates were washed with acetone and annealed under vacuum condition ( $<10^{-7}$  torr) at  $900^{\circ}\text{C}$  for 10 hours. The samples were cooled down to room temperature in furnace and washed with aqua regia to remove impurity on the surface of the palladium samples. After that, some samples were covered with MgO or Al by ion beam sputtering, some samples were electroplated by copper or platinum, and the others had no films. The aim of the surface modification is to reduce the rate of deuterium gas release from the vacuum side of the palladium.

### 3. Data Analysis

D/Pd is estimated by the following procedure. Equation of state for the upper part of the electrolyte side is expressed as;

$$p_{O_2} V_{cell} = N_{O_2} \kappa T_{cell} \quad (1)$$

where *cell* denotes upper part of the electrolyte side and  $\kappa$  is the Boltzmann constant. The number of deuterium atoms absorbed into the palladium is given by

$$N_{D_{in}} = 2N_{D_2} = 4N_{O_2} = \frac{4V_{cell} p_{O_2}}{\kappa T_{cell}} \quad (2)$$

As the vacuum chamber is evacuated by the pumping speed of  $S$ , an equation of mass balance is obtained as

$$\frac{d}{dt} (N_{vac} \kappa T_{vac}) = V_{vac} \dot{p}_{vac} + p_{vac} S \quad (3)$$

where denotation of *vac* means the vacuum side. Almost of the gas in the vacuum chamber is composed of  $D_2$  gas, then time derivative of the number of deuterium atoms is given by

$$\dot{N}_{D_{out}} = 2 \frac{V_{vac} \dot{p}_{vac} + p_{vac} S}{\kappa T_{vac}} \quad (4)$$

Combining equation (2) and (4), we obtain the following equation;

$$D/Pd = \frac{M_{Pd}}{\omega_{Pd} N_{AB}} \left[ \frac{4V_{cell} p_{O_2}}{\kappa T_{cell}} - 2 \int_{t_0}^t \frac{V_{vac} \dot{p}_{vac} + p_{vac} S}{\kappa T_{vac}} dt \right] \quad (5)$$

where  $N_{AB}$ ,  $M_{Pd}$ ,  $\omega_{Pd}$  and  $t_0$  denote Avogadro number, mass number of palladium, mass of the palladium sample and the time that  $D_2$  gas begins to release out, respectively.

Next, we describe the excess heat analysis. Assumption of the analysis is that we consider only steady state and dissipated heat from the apparatus is estimated by Pd/H<sub>2</sub>O system.

Heat balance equation of the apparatus is expressed as

$$P_{ex} + P_{in} - P_{out} - P_{diss} - \Delta \dot{H}_{gas} + \Delta \dot{H}_{abs} - \Delta \dot{H}_{des} = 0, \quad (6)$$

where *ex*, *in*, *out*, *diss*, *gas*, *abs* and *des* mean excess, input, output, dissipation, gas release, absorption and desorption. The terms of gas release, absorption and desorption of deuterium are negligible if we use typical experimental parameters. Therefore excess heat is calculated by

$$P_{ex} = \dot{m}_w c_w (T_{out} - T_{in}) - IV + \int_A h (T_{sol} - T_{room}) dA \quad (7)$$

where *IV* is the input power, *m<sub>w</sub>* and *c<sub>w</sub>* are mass of the water and specific heat, *h* is the heat transfer coefficient and *A* means the whole surface of the apparatus.

#### 4. Results and Discussion

Figure 2 shows an example of D/Pd analysis. In this case, the sample has no surface film. Since the deuterium atoms absorbed on the surface of the electrolyte side of the palladium do not reach the opposite surface, deuterium gas is not released at the early period of the experiment. D/Pd increases gradually and attains to 0.8 at the time of  $1 \times 10^5$  sec. This results indicate that D/Pd reaches about 0.8 even for the palladium sample without surface barrier on the vacuum side of it.

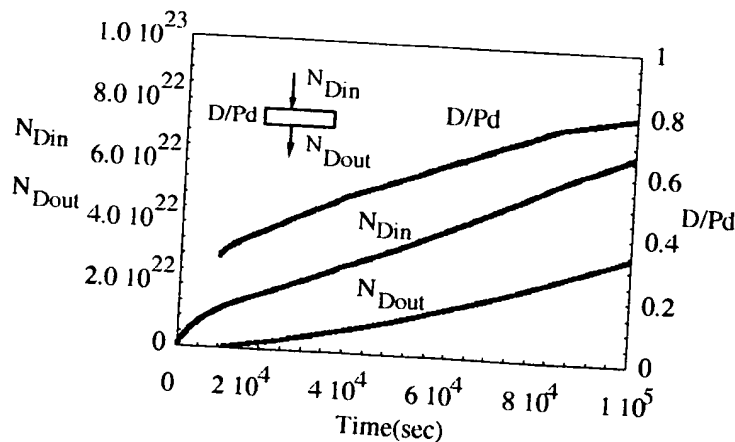


Fig.2 D/Pd Analysis

Variation of deuterium behavior depending on palladium samples is shown in Fig.3. Pressure(electrolyte) lines mean the pressures of the upper side of the electrolyte side which is proportional to the quantity of deuterium into palladium, and Pressure(vacuum) lines the vacuum side proportional to time derivatives of the number of released deuterium atoms.

These samples, EV29 and EV34, are prepared by the same procedure; they are derived from the same lot and the method of annealing and etching is all the same. However, it is easy to realize that absorption and desorption of deuterium is entirely different. It suggests that absorption and desorption behavior of deuterium is greatly influenced by unspecified factors; metallurgical conditions such as impurity and defects in palladium.

# Nuclear Physics Approach

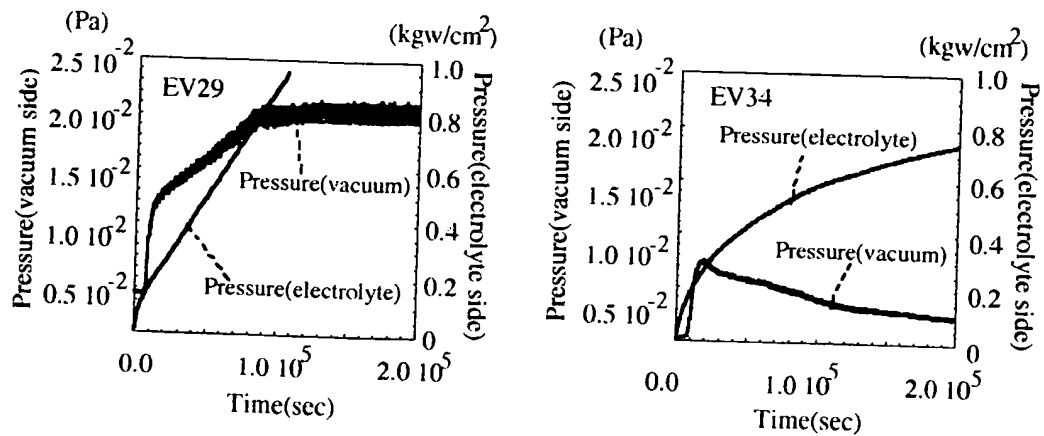


Fig.3 Variation of Absorption and Desorption of Deuterium

An example of long period x-ray emission and its energy spectrum is demonstrated in Fig.4. This sample has Al thin film ( $\sim 400\text{\AA}$ ). Upper figure indicates time variation of x-ray (see Fig.1) located in the vacuum chamber. X-ray count rate reaches over 10 times larger than that of background. We can see that x-ray emission lasts for a long period; more than 1 day. It was a successful example in the meaning that we could make long-term continuous nuclear reactions occur in the Pd-D<sub>2</sub> system.

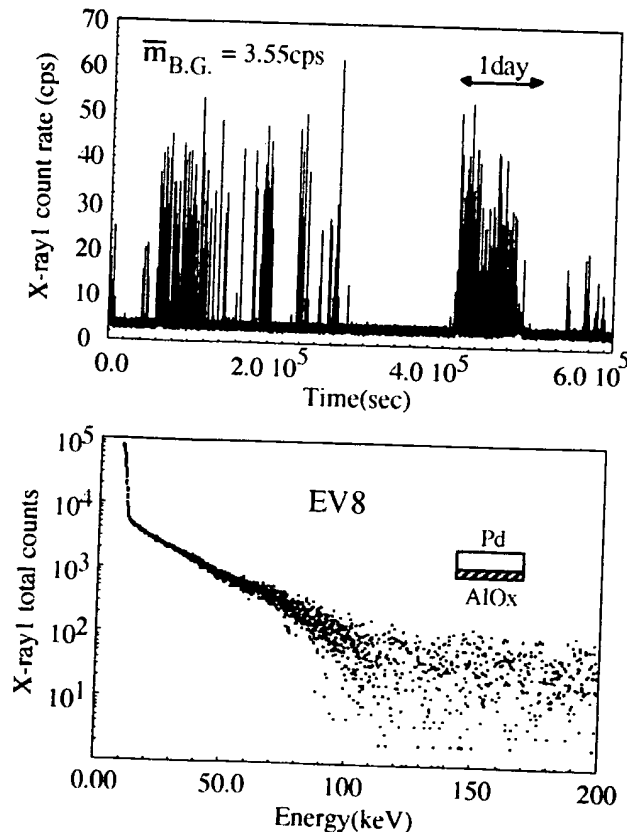


Fig.4 Long-term X-ray Emission and its energy spectrum

An energy spectrum of the x-ray emission for about a week is plotted in the lower figure. X-ray energy distributes continuously as shown in the figure. However, at present, the authors cannot explain consistently why such an energy spectrum was obtained. It is an important future work.

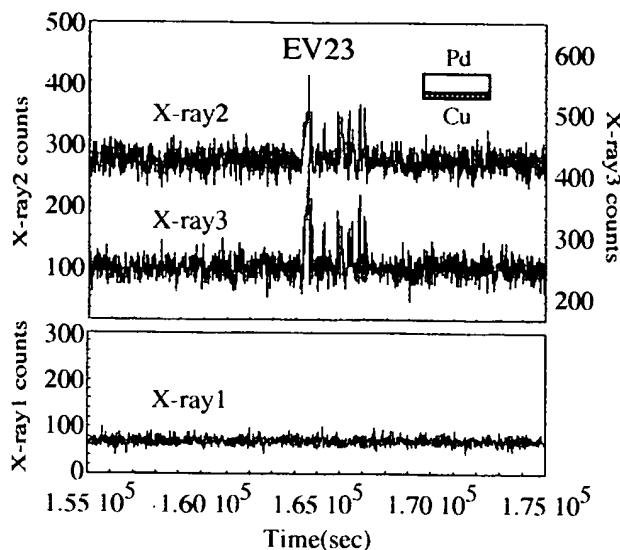


Fig.5 Simultaneous detection by upper side x-ray detectors

Next example shows simultaneous detection by the upper side of x-ray NaI counters(Fig.5). The sample EV23 is electroplated by copper. Coincidence of x-ray2 and x-ray3 is very good, on the other hand, counts of x-ray1 are almost equal to those of background. In the 14 out of 40 cases, we observed this kind of x-ray emissions. Judging from the experimental results, nuclear reactions must occur on the electrolyte side of the palladium. For the thickness of palladium is 1mm, reduction rate of x-rays through the palladium is larger than that through the D<sub>2</sub>O(4cm).

Figure 6 illustrates the correlation between excess heat and the other parameters. In this case(EV39), we use a palladium sample covered with MgO thin film. At low current, any excess heat cannot be seen. However, after making the applied current up to 3A (current density : 1.5A/cm<sup>2</sup>), we can observe a clear excess heat generation. Input power is about 40W when current of 3A is applied, therefore excess heat we obtain is a few percentages of the input power.

Comparison of heat distribution between EV39(excess heat generation) and EV40(no excess heat) is shown in Figure 7. In both cases, the other experimental conditions are the same except the palladium sample treatment. MgO thin film is formed on the vacuum side of EV39, and Fe thin film on the electrolyte side of EV40. It is clear that mean value and deviation of excess heat of EV39 are larger than those of EV40.

Up to now, several times we observed excess heat generation, however, we could not see any relations between excess heat generation and x-ray emission. In some cases, excess heat generation seems to be related with pressure in vacuum chamber; it corresponds to time derivatives of the number of deuterium atoms. As for neutron measurement, a clear emission has not been observed during the experiments using this apparatus. Nevertheless, the authors consider that we must investigate furthermore to conclude correlation between excess heat and nuclear products.

# Nuclear Physics Approach

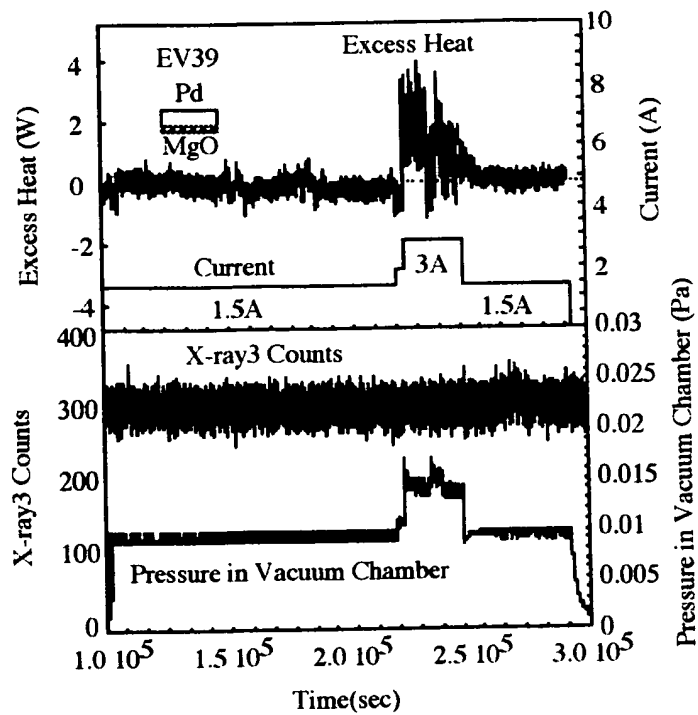


Fig.6 Correlation between Excess Heat and the other parameters

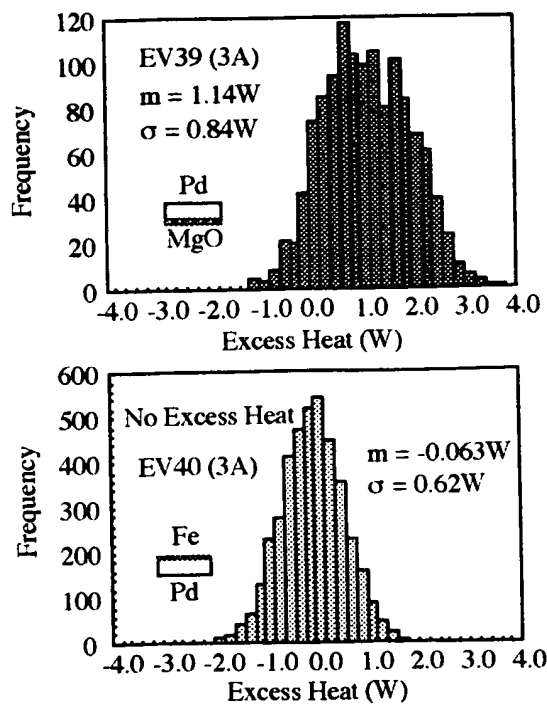


Fig.7 Excess Heat Distribution



If we assume a few MeV energy release by an event, obtained excess heat corresponds to the order of  $10^{12}$  events/sec. On the other hand, observed x-ray emissions range from  $10^2$  to  $10^4$  events/sec (calibrated by an Am-241 source). Judging from these results, we might consider that excess heat and x-rays are generated by different nuclear reactions. In our previous paper<sup>3</sup>, it was suggested that x-ray and neutron are generated by different reactions. Therefore it seems that we should not exclude the possibility that various nuclear reactions occur and produce various nuclear ashes.

### 5. Concluding Remarks

A new type of experimental apparatus for simultaneous measurement was developed. Absorption and desorption behavior of deuterium in palladium was greatly changed by the unspecified conditions of the palladium. X-ray emission lasting for long term and excess heat generation was observed. Up to now, correlation between excess heat and x-rays has not been clear under our experimental condition. Nevertheless, we must investigate furthermore to conclude correlation between excess heat and nuclear products.

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# DETECTION OF ANOMALOUS ELEMENTS, X-RAY AND EXCESS HEAT INDUCED BY CONTINUOUS DIFFUSION OF DEUTERIUM THROUGH MULTI-LAYER CATHODE(Pd/CaO/Pd)

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## **Abstract**

A new type of experimental apparatus is developed to induce nuclear reactions by continuous diffusion of deuterium. Ti atoms, which cannot be explained by contamination, were detected on the surface where deuterium atoms passed through on Pd cathodes after electrolysis. A multi-layer cathode(Pd/CaO/Pd) is introduced based on an EINR(Electron Induced Nuclear Reaction) model. Excess heat generations and x-ray emissions were observed for all the cases we tried by the multi-layer cathodes.  $^{57}\text{Fe}/^{56}\text{Fe}$  ratio of Fe atoms detected on the multi-layer cathodes is anomalously large than natural  $^{57}\text{Fe}/^{56}\text{Fe}$ .

## **1. Introduction**

Beginning in 1993, we have researched "cold fusion" phenomena to investigate its potentiality as a new energy source. At first, we performed gas-loading experiments and suggested that the high diffusion velocity of deuterium, in addition to a high D/Pd ratio, is an important factor for causing nuclear reactions in solids<sup>(1)-(3)</sup>. The authors analyzed electrolyzed Pd samples by a variety of methods<sup>(4)</sup>, and we conjectured that impurities in Pd play essential roles to induce nuclear reactions. The foregoing ideas result in the assumption that necessary conditions to induce nuclear reactions in solids are as follows:

- (i) high D/Pd
- (ii) enough diffusion flux of deuterium
- (iii) the existence of a third element except Pd and deuterium.

A new type of experimental apparatus was developed to induce continuous diffusion under high D/Pd conditions, in which the conditions (i) and (ii) were satisfied<sup>(5)</sup>. A multi-layer cathode composed of a Pd sheet, Pd and CaO complex layer, and Pd thin layer is developed to meet the condition (iii). Ca is introduced into Pd cathode based on an Electron-Induced Nuclear Reaction(EINR) model<sup>(6)</sup>.

In this paper, experimental results using the continuous diffusion apparatus with both a normal Pd sample and the multi-layer cathodes is described.

## **2. Experimental**

Fig.1 shows a cross sectional view of the continuous diffusion experimental apparatus<sup>(3),(6)</sup>. An electrolyte of 1M LiOD/D<sub>2</sub>O and a vacuum chamber are separated by a Pd plate with an O-ring gasket(KARLEZ). Deuterium atoms are loaded by electrochemical potential into one side of the Pd sample and released from the other side. With this composition, it is possible to control the state of diffusion of deuterium by applied current or pressure of the vacuum side.

Excess heat is estimated by the flow calorimetry method. Heavy water (up to 99.9%) and LiOD (up to 99%) are provided by ISOTEC Inc. Electrolyte side of the apparatus consists of a cathode of Pd plate(25x25x1mm; 99.9% Tanaka Kikinzoku Kogyo K.K.), a circular shape of anode of platinum mesh(φ0.5mm; 99.98% Nilaco Co.), a recombiner and a cooling pipe for measurement of excess heat generation.

Pressures of the upper part of electrolyte and vacuum chamber are monitored by a pressure gauge and a BA gauge, respectively. At the beginning of electrolysis, argon gas is filled up at 1 atm in the upper space of the electrolyte.

The recombiner is prepared by electroplating Pt mesh in  $H_2PtCl_6$  solution. In this apparatus, increase of pressure of the upper part of electrolyte corresponds to absorbed deuterium. Recombiner efficiency is calculated by the analysis of  $D_2$  and  $O_2$  gases in the upper part of the electrolyte using a quadrupole mass spectrometer(AQA-360, ANELVA). Furthermore, temperature of the recombiner is always monitored to make sure that the recombiner works. We confirm that the recombiner works by the temperature and gas analysis. The usual recombiner efficiency that we measured was  $> 99\%$ .

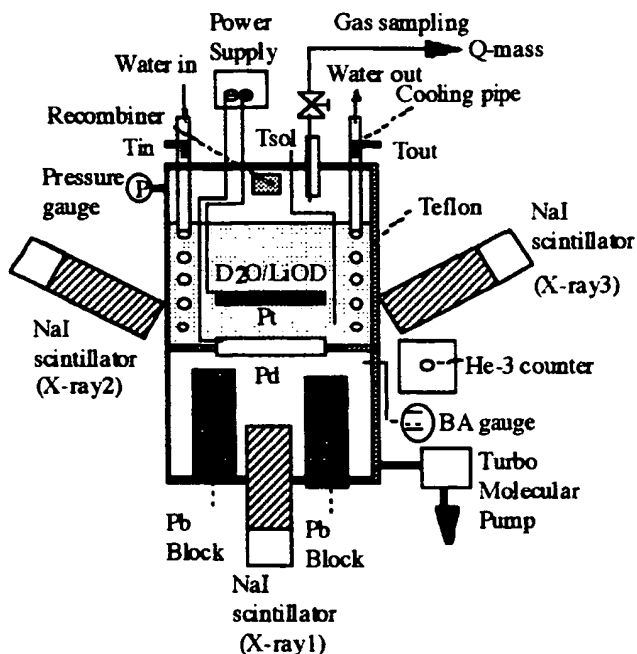


Fig.1 Continuous Diffusion Experimental Apparatus

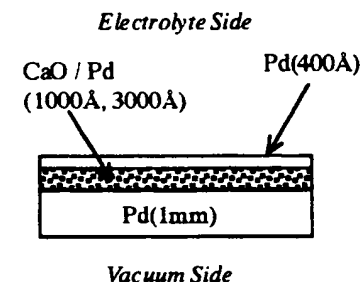


Fig.2 Structure of a Multi-Layer Cathode

The cooling pipe is electroplated with gold( $10\mu m$  thickness) because Au is resistant to alkaline solution and it has good thermal conductivity. Flow rate of coolant (pure water) is always measured at 2 points: inlet and outlet sides. Two thermocouples each are provided to measure the inlet and outlet temperatures of the water. The Solution, gas, recombiner and environmental temperatures are measured and consistency among these temperatures is always checked. The cell of the electrolyte side of the apparatus is made of Teflon. All experimental parts such as electric wires in the electrolyte side are coated with Teflon. The apparatus and measuring systems are located in a clean-room where temperature and humidity are always controlled at constant levels ( $23\pm 1^\circ C, 40\pm 5\%$ ). Description on our nuclear measurement is provided in Ref.5 and 6. The procedure for sample preparation is as follows. Pd plates were washed with acetone and annealed under vacuum condition ( $< 10^{-7}$  torr) at  $900^\circ C$  for 10 hours. The samples were cooled down to room temperature in furnace and washed with aqua regia(D) for 100sec to remove impurity on the surface of the Pd samples. In the case of preparing a multi-layer cathode shown in Fig.2, the sample was covered with a complex layer which consisted of CaO and Pd. It was formed by simultaneously sputtering of CaO and Pd with Ar ion beam. Thickness of the layer was set from  $1000\text{\AA}$  to  $3000\text{\AA}$ . A thin Pd layer( $400\text{\AA}$ ) was formed on it by ion beam sputtering to avoid dissolving CaO into  $D_2O/LiOD$  solution. The complex layer is put at near surface of the electrolyte side because we assume that nuclear reactions occur at the near surface.

### 3.Results and Discussion

Figure 3 shows the appearance of a normal Pd sample after experiments. Excess heat of about 1W lasted for 1 day in the case of the sample, although x-ray and neutron were not detected. The black circle of the electrolyte side (surface A) corresponds to the

place where deuterium atoms passed through; it is the shape of the Pt anode. Surface B is the place where no deuterium atoms pass through. Similar black circles corresponding to the shape of the anode are observed on multi-layer cathodes. Contrarily, the appearance of the vacuum side of the Pd is the same as before experiments, independent of the deuterium passage.

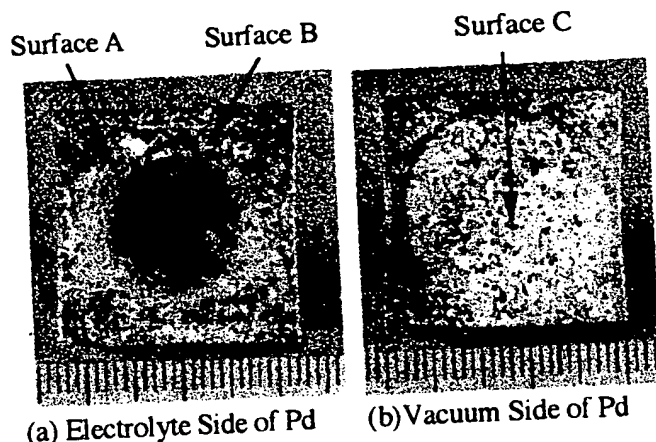


Fig.3 The Electrolyte and Vacuum Sides of Pd

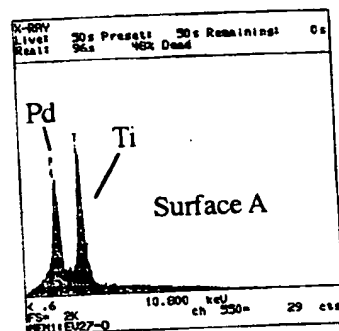


Fig.4 EDX Spectrum for Surface A

Figure.4 show the results of EDX(Energy Dispersive X-ray Spectrometry) for the surface A. Ti is clearly seen in the EDX spectrum. The Ti element was confirmed by WDX(wavelength dispersive X-ray spectroscopy), AES(Auger Electron Spectrometry), XPS(X-ray Photoelectron Spectrometry) and ICP-MS(Inductively Coupled Plasma Mass Spectrometry). Those results are consistent with each other. On the other hand, no element except Pd is detected on the surface B and C.

Ti element on the black circle(surface A) was estimated  $21\mu\text{g}$  by the ICP-MS. Maximum quantity of Ti contained in LiOD/D<sub>2</sub>O solution and a Pt anode can be estimated  $3.3\mu\text{g}$ . The LiOD/D<sub>2</sub>O solution was kept for 3 days in the cell before analysis. Mass of Ti in a whole Pd cathode before electrolysis is about  $19\mu\text{g}$ . Therefore, the maximum total mass of Ti is  $22.3\mu\text{g}$  which is the same order of the increased mass of Ti on the surface A.

ICP-MS analysis shows that total mass of Pt or Cu in the electrolyte, Pt and Pd is much larger than Ti. However, Pt and Cu was not detected on the surface A. It is very difficult to consider that only Ti elements were concentrated on the electrolyte surface where deuterium atoms passed through by an ordinary physicochemical process. After experiments, we keep the Pd samples in a desiccator in the clean room. The possibility of Ti being deposited on the Pd after the experiments is extremely low. Therefore the authors consider that Ti is formed by certain nuclear process in solids.

The experimental results using the multi-layer cathodes are summarized in Table 1. All the samples except EV54 belonged to the multi-layer cathodes, although thicknesses of CaO/Pd layers were different. In the table,  $\sigma_x$  denotes standard deviation of the background for each detector. We judge that excess heat is generated only if it exceeds  $3\sigma_H$  of background(Pd-H<sub>2</sub>O system). The excess heat is evaluated by the increase of coolant temperature. Details of excess heat and x-ray analyses are described in Ref.5 and 6.

Both excess heat generation and x-ray emission were observed for all the multi-layer cathodes as shown in Table 1. Input power ranges from 20W to 40W. X-ray emissions were simultaneously detected by X2 and X3 counters located at the upper side of the Pd. Maximum deviations from mean values of X2 and X3 were shown in the table. Neutron emission of  $22\sigma_H$  was observed only in the case of EV50.

Elements on the surface A of multi-layer cathode after experiments detected by EPMA(Electron Probe Micro Analysis) are shown in the table. The electrolyte, Pt anode, and Pd contain much more Fe, Cu, Si than Ti. The maximum quantities of them in the

apparatus easily exceed the quantities of detected elements on the cathode. Several papers<sup>(7)-(9)</sup> report isotope shifts. We apply SIMS(Secondary Ion Mass Spectroscopy) to the detected Fe and show that Fe, at least, can not be explained by contamination.

Mass number ratios (mass57/mass56) are also shown in the table. For some samples, SIMS analyses were performed at several points. The values of the table were obtained by integrating depth profiles. It is noticeable that all the values of mass57/mass56 on multi-layer cathodes are distributed from 0.036 to 0.66; they are larger than 0.023 which is obtained by Fe standard material.

We should consider the following species as the candidates of mass 54, 56, 57 and 58, because a variety of elements on the cathodes are detected by SIMS.

mass54 :  $^{54}\text{Fe}$ ,  $^{54}\text{Cr}$ ,  
mass57 :  $^{57}\text{Fe}$ ,  $^{56}\text{FeH}$ ,  $^{40}\text{Ca}^{16}\text{OH}$   
mass56 :  $^{56}\text{Fe}$ ,  $^{40}\text{Ca}^{16}\text{O}$ ,  
mass58 :  $^{58}\text{Fe}$ ,  $^{58}\text{Ni}$ ,  $^{56}\text{FeD}$ ,  $^{40}\text{Ca}^{16}\text{OD}$

Examples of Depth profile of those mass numbers are shown in Fig.5 and 6. SIMS measurements were performed by MST(Foundation for Promotion of Material Science and Technology of Japan) with Physical Electronics 6600. We see that secondary ion intensity and mass number ratio of EV53 behave anomalously. Especially, mass57/mass56 reaches 0.8, although natural  $^{57}\text{Fe}/^{56}\text{Fe}$  is 0.023. On the other hand, secondary ion intensity and mass number ratio of EV54 are distributed uniformly, which is a Pd cathode used in a light water experiment. Depth profiles of Fe standard material are similar to those of EV54.

Table 1 Summary of Multi-Layer Cathode Experiments

Sample	Structure of Cathode	Excess Heat	X-ray	Detected Elements	mass57/mass56
EV50	Pd 400Å CaO/Pd 1000Å	Generation (max 3.2W)	X1 : Background X2,X3: Simultaneous Detection X2(6.6σ <sub>X2</sub> ),X3(6.2σ <sub>X3</sub> )	Si, Fe, Cu	0.036 0.038 0.065
EV51	Pd 400Å CaO/Pd 3000Å	Generation (max 1.5W)	X1 : Background X2,X3: Simultaneous Detection X2(6.4σ <sub>X2</sub> ),X3(7.8σ <sub>X3</sub> )	Fe, Cu	0.24
EV52	Pd 400Å CaO/Pd 1000Å	Generation (max 1.9W)	X1 : Background X2,X3: Simultaneous Detection X2(7.6σ <sub>X2</sub> ),X3(8.8σ <sub>X3</sub> )	Fe, Cu	0.66 0.26 0.22 0.29
EV53	Pd 400Å CaO/Pd 3000Å	Generation (max 1.8W)	X1 : Background X2,X3: Simultaneous Detection X2(14.4σ <sub>X2</sub> ),X3(16.6σ <sub>X3</sub> )	Fe, Cu	0.45 0.37 0.66
EV61	Pd 400Å CaO/Pd 1000Å	Generation (max 2.3W)	X1 : Background X2,X3: Simultaneous Detection X2(6.4σ <sub>X2</sub> ),X3(6.7σ <sub>X3</sub> )	Fe, Au, Cu	0.097
EV54	Pd only (light water)	None (<3σ <sub>H</sub> )	X1,X2,X3: Background	Fe	0.030 0.030
Fe Standard Material	—	—	—	—	0.023 0.023 0.022

Let us consider the relation between mass57/mass56 and  $^{57}\text{Fe}/^{56}\text{Fe}$ . If we substitute 0.036 of EV50 into mass57/mass56, we obtain

$$\frac{\text{mass57}}{\text{mass56}} = \frac{^{57}\text{Fe} + ^{56}\text{FeH} + ^{40}\text{Ca}^{16}\text{OH}}{^{56}\text{Fe} + ^{40}\text{Ca}^{16}\text{O}} = \frac{^{57}\text{Fe}}{^{56}\text{Fe} + ^{40}\text{Ca}^{16}\text{O}} + \frac{^{56}\text{FeH}}{^{56}\text{Fe} + ^{40}\text{Ca}^{16}\text{O}} + \frac{^{40}\text{Ca}^{16}\text{OH}}{^{56}\text{Fe} + ^{40}\text{Ca}^{16}\text{O}} = 0.036$$

where all the symbols of the elements mean secondary ion intensity. Every term is positive, therefore

$$\frac{{}^{56}\text{FeH}}{{}^{56}\text{Fe}+{}^{40}\text{Ca}^{16}\text{O}} < 0.036 \frac{{}^{40}\text{Ca}^{16}\text{OH}}{{}^{56}\text{Fe}+{}^{40}\text{Ca}^{16}\text{O}} < 0.036$$

$$\frac{\text{mass}57}{\text{mass}56} < \frac{{}^{57}\text{Fe}}{{}^{56}\text{Fe}+{}^{40}\text{Ca}^{16}\text{O}} + 0.036 + 0.036 < \frac{{}^{57}\text{Fe}}{{}^{56}\text{Fe}} + 0.072$$

$$\therefore \frac{{}^{57}\text{Fe}}{{}^{56}\text{Fe}} > \frac{\text{mass}57}{\text{mass}56} - 0.072$$

If we assume that the effects of Ca are the same as EV50, then  ${}^{57}\text{Fe}/{}^{56}\text{Fe}$  of EV52 is larger than 0.15.  ${}^{57}\text{Fe}/{}^{56}\text{Fe}$  of EV53 is larger than 0.30.

According to the Table1, we may say that  ${}^{57}\text{Fe}/{}^{56}\text{Fe}$  ratios of EV51, 52 and 53 are anomalously large. The large  ${}^{57}\text{Fe}/{}^{56}\text{Fe}$  ratio and the anomalous behavior of SIMS data in Fig.5 indicate that the detected Fe atoms are not produced by normal chemical process. Therefore it is strongly suggested that nuclear reactions occur on the multi-layer cathodes.

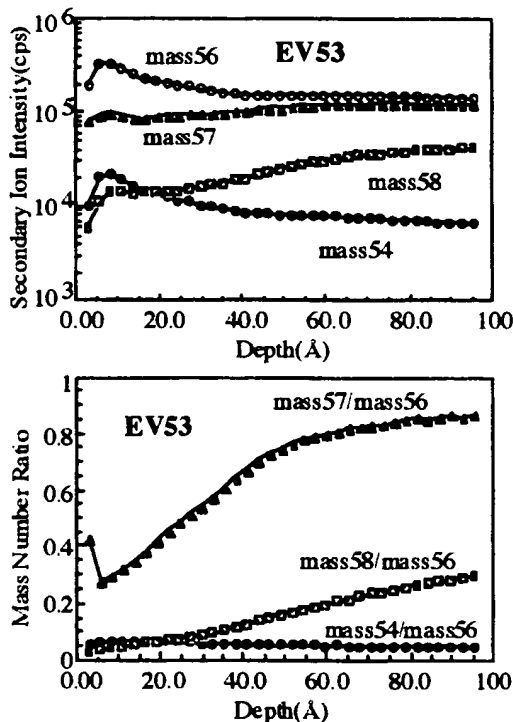


Fig.5 Depth Profiles of Mass Numbers Related to Fe isotopes (EV53: multi-layer cathode)

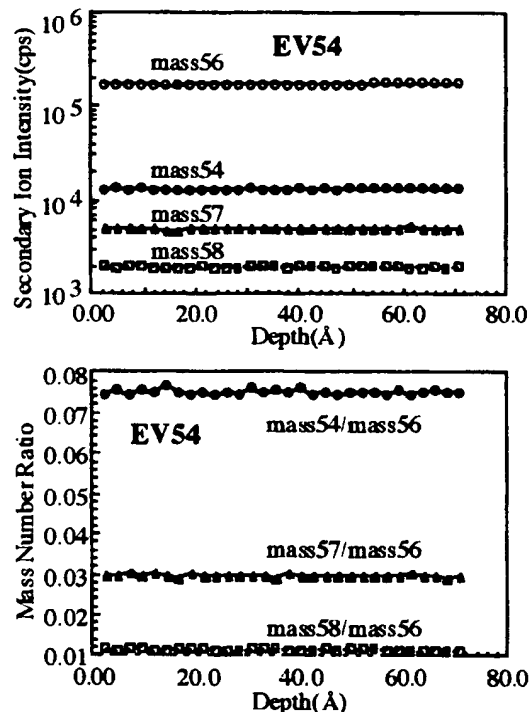


Fig.6 Depth Profiles of Mass Numbers Related to Fe isotopes (EV54: Pd cathode, light water)

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## II. INHIBITED DIFFUSION DRIVEN SURFACE TRANSMUTATIONS

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This paper is the second of a set of three papers dealing with the role of coherent partitioning as a common element in Low Energy Nuclear Reactions (LENR), by which is meant cold-fusion related processes. This paper discusses the first step in a sequence of four steps that seem to be necessary to explain Iwamura 2- $\alpha$ -addition surface transmutations. Three concepts are examined: salt-metal interface states, sequential tunneling that transitions  $D^+$  ions from localized interstitial to Bloch form, and the general applicability of 2-dimensional vs. 3-dimensional symmetry hosting networks.

### 1. Introduction

Iwamura et al.<sup>1</sup> have developed an LENR process that converts surface Cs atoms into surface Pr atoms using deuterium permeation through a specially structured, largely metal assembly. The assembly consists of a Pd substrate on which are deposited four layers of 2-nm CaO overcoated with 8-nm Pd, plus one CaO layer overcoated with a 40-nm Pd inflow layer. The Iwamura assembly is used as a permeable barrier between a  $D_2$ -pressurized volume and a vacuum volume.

### 2. Envisioned Process

The author speculatively models the process as follows. At a CaO-Pd interface, a diffusing deuteron is converted into a coherently partitioned, nuclearly reactive Bloch form  $D_{\text{Bloch}}^+$ .<sup>2</sup> Multiple Bloch deuterons organize themselves into a many-body Bloch-ion subsystem. Independent subsystems form on different CaO crystallites as shown in Fig.1. In these subsystems Bloch deuterons fuse, creating Bloch helium ions  ${}^4\text{He}_{\text{Bloch}}^{2+}$ . In a second step two  ${}^4\text{He}_{\text{Bloch}}^{2+}$  fuse, creating a product nucleus  ${}^8\text{Be}_{\text{Bloch}}^{4+}$ , which is stabilized by being coherently partitioned. The  ${}^8\text{Be}_{\text{Bloch}}^{4+}$  is energized by increases in coherent partitioning in a process that uses nuclear energy to mobilize the Be ion. The  ${}^8\text{Be}_{\text{Bloch}}^{4+}$  nucleus spreads out beyond the hosting CaO-metal interface, spreading along accessible Pd-crystallite boundaries, including the interfaces between metal and gas, and metal and vacuum. A Cs atom protruding above the mean surface of the Pd metal is overlapped and infiltrated by the  ${}^8\text{Be}_{\text{Bloch}}^{4+}$ . The Cs nucleus combines with and absorbs the  ${}^8\text{Be}_{\text{Bloch}}^{4+}$  nucleus, and becomes a Pr atom. Both the formation of Bloch helium and the transmutation of surface Cs into Pr are exothermic processes.

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### 3. Primary Challenge

The core question is: Why does Bloch deuterium form within the CaO-metal interfaces? My guess is that within the interface layer the Bloch deuterium configuration has lower free energy than such discrete particles as  $\text{D}^+$ ,  $\text{D}^-$ , or adsorbed free-radical D atoms. This is generally not the case at the metal-gas and metal-vacuum interfaces, where electron-neutralized surface  $\text{D}^+$  ions must be in excited states to be of Bloch configuration.<sup>3</sup> Bulk CaO is a face centered NaCl lattice with a large Gibbs free energy of  $-898 \text{ kJ/mol}$ . One might imagine that a CaO crystal, with its alternating positive ion, negative ion structure, when interfaced with deuterided Pd could host a fraction of a  $\text{D}^+_{\text{Bloch}}$  ion adjacent to each interface  $\text{O}^{2-}$  ion and a fraction of a  $\text{D}^-_{\text{Bloch}}$  ion adjacent to each interface  $\text{Ca}^{2+}$  ion. Each fractionally distributed Bloch deuteron and its fractionally distributed neutralizing electron charge would be in Bloch-function configuration. In Fig. 2, the CaO lattice determines the Bloch periodicity, while the interface boundary electrons of the more plastic metal adjust to ensure unit cell neutrality. The result is a partitioned  $\text{D}^+$  ion with a local maximum in density within each of a large number of non-self-trapping potential wells.<sup>4</sup> The essential step that transitions localized  $\text{D}^+$  into a delocalized Bloch configuration presumably occurs within these CaO-Pd interface layers, as discussed later. Bloch deuteron pairs

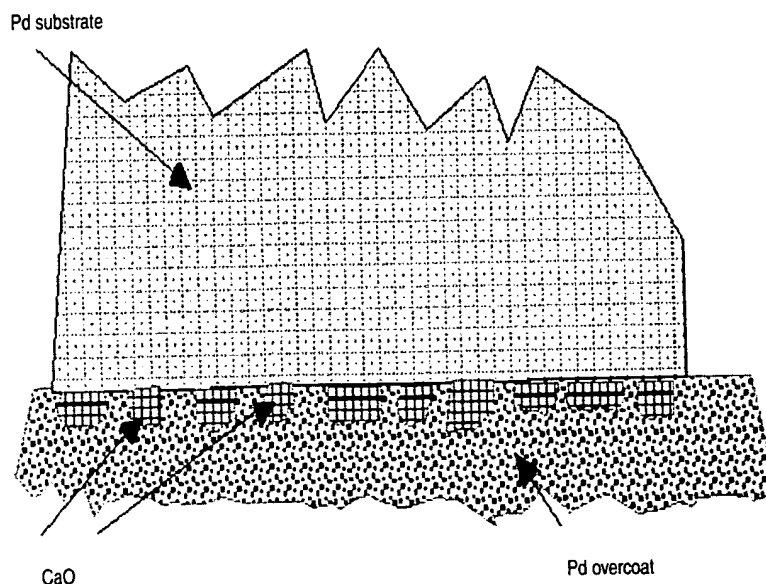


Figure 1. Multi-crystallite modeling of Iwamura reactor plate, showing contacts between Pd substrate, sputtered CaO deposition layer, and sputtered Pd metal overcoat. The CaO is shown as a discontinuous set of ionic nano-crystals.



then convert into wavelike  ${}^4\text{He}_{\text{Bloch}}^{2+}$ , which participate in Step 2 of the Iwamura process.

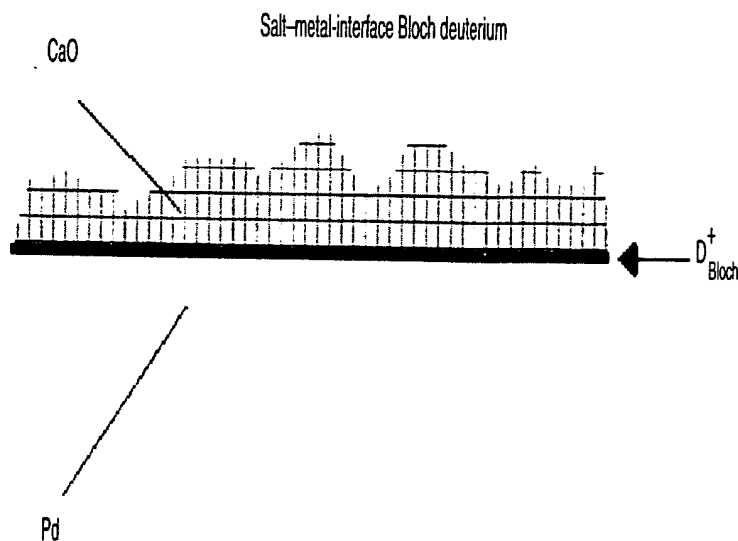


Figure 2. Fine scale view of nano-portion of interface between ionic crystallite CaO and transition metal Pd, as fabricated for the Iwamura reactor. Arrow points to the location of the hypothesized  $\text{D}_{\text{Bloch}}^{+}$  deuteron. The  $\text{D}_{\text{Bloch}}^{+}$  ion is a nucleus with 2-dimensional symmetry matching the template provided by the CaO crystallite, and is neutralized (dressed) by the metal's electron Fermi sea.

#### 4. Impact on Cold Fusion

The Iwamura et al.  $\alpha$ -addition transmutation discoveries are the first of two new discoveries that clarify the role of coherent partitioning and Bloch-sensitivity in LENR processes. The Iwamura program started as a Mitsubishi Heavy Industries research study exploring anomalous Fleischmann-Pons (F-P) emissions. Mitsubishi support allowed the science team to evolve their program into use of permeation as a means of avoiding electrolysis contamination worries. The Iwamura protocol is a non-electrolytic deuterium-palladium process by which surface Cs atoms have been repeatedly transmuted into surface Pr atoms. The Iwamura team has identified a new class of transmutations. The archetype is  ${}^{133}\text{Cs} + 2\alpha \rightarrow {}^{141}\text{Pr}$ , which is a type of

$\alpha$ -addition transmutation through Pd metal by exposure of a Pd plate reactor to  $\text{D}_2$  gas. The side of the plate is pumped, and the reactions take place without the use of a Pd foil support plate or a layer of diffusion-inhibiting layers of CaO salt, separating the Pd metal, and topped with a coating of Cs atoms is deposited. The process occurs during week-long experiments measured by X-ray photoelectron spectroscopy and twice during imposition of small CaO crystals, permeating through the Pd plate. The process is for D-permeation, or if it is not.

The Iwamura process uses the F-P process with  $x \equiv D/\text{Pd} > 0.85$ . The process uses pressures  $>$  several kbar from exothermic nuclear reactions in a manner that heats a hot place at less than standard conditions.

The Iwamura process is the Iwamura process or relatively low  $\text{D}_2$  chemical potential. CaO crystals can effect the transmutation of a high  $Z$  nucleus in the addition of two alphas (see Table 1. Challenges 2-4

#### 5. Low $\text{D}_{\text{Bloch}}^{+}$ Concentration

Let us consider Iwamura's  $\text{D}_{\text{Bloch}}^{+}/\text{Pd}$  ratio. As determined by the CaO salt, it is unusually suitable for ionic salt like NaCl, a salt that its ordered form provides the template for wave-charge cloud that converts

" $\alpha$ -addition transmutation." In the Iwamura process deuterium is forced to diffuse through Pd metal by exposing the front surface of the previously described special Pd plate reactor to  $D_2$  gas at 1 atm pressure at  $70^\circ C$ , while the gas from the back side of the plate is pumped away using a vacuum pump. The transmutations do not take place without the use of a complex plate reactor. The specified design describes a Pd foil support plate of 0.1 mm thickness, on which are sputter-deposited a set of diffusion-inhibiting layers. Inside the plate there are five sputter-deposited 2-nm layers of CaO salt, separated from each other by sputter-deposited 8-nm layers of Pd metal, and topped with a 40-nm layer of sputter-deposited Pd. A sub-monolayer coating of Cs atoms is deposited on the top surface. The transmutation of Cs into Pr occurs during week-long permeation runs. Cs and Pr surface concentrations are measured by X-ray photoelectron spectroscopy (XPS) before and after permeation, and twice during imposed run interruptions. The salt layers, likely in the form of small CaO crystals, partially block the deuterium atoms as they work their way through the Pd plate. Transmutation is not observed if H-permeation is substituted for D-permeation, or if the CaO layers are not included in the plate structure.

The Iwamura process differs from F-P methods in that it avoids electrolysis. The F-P process uses overvoltage electrolysis to create a  $PdD_x$  interstitial deuteride with  $x \equiv D/Pd > 0.85$ . To achieve  $x > 0.85$  by equilibrium chemistry requires  $D_2$  pressures  $>$  several kbar.<sup>5</sup> On the other hand, both processes release nuclear energy from exothermic nuclear reactions, and transfer the released nuclear energy in a manner that heats a hosting metal lattice. Step 1 in the Iwamura process takes place at less than standard deuterium chemical potential.

The Iwamura process imposes challenges to cold fusion modelers. To model the Iwamura process one must explain: (1) why nuclear energy release occurs at relatively low  $D_2$  chemical potential, (2) how a process made possible by interior CaO crystals can effect transmutation on a distant metal surface, (3) how the transmutation of a high  $Z$  nucleus can occur, and (4) why the transmutation involves addition of two alphas instead of one. This paper is concerned mainly with Challenge 1. Challenges 2-4 are addressed in Paper III.

## 5. Low $D_{\text{Bloch}}^+$ Concentration Problem

Let us consider Iwamura's success in achieving nuclear effects at relatively low  $D_{\text{Bloch}}^+/Pd$  ratio. As described above, it is suggested that the periodic structure provided by the CaO salt crystallites helps create a one layer thick interface volume unusually suitable for hosting coherently partitioned Bloch deuterons. CaO is an ionic salt like NaCl, and has a large negative Gibbs free energy, which suggests that its ordered form is not easily altered. In this speculation, the CaO forms the template for wave-like deuterium, and the metal's electrons forms the negative charge cloud that converts a positive ion into the neutral "atom" form.

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## 6. Initial Step in Coherent Partitioning

A relatively simple process can be envisioned by which the Iwamura plate reactor creates coherently partitioned deuterons. An incident  $D_2$  gas molecule strikes the Pd top surface and dissociates into two adsorbed D atoms. Each adsorbed D atom enters the Pd top surface by tunneling into an interior self-trapping octahedral site within the fcc lattice. The permeating deuteron then diffuses through the top sputtered Pd layer in response to an internal deuteron concentration gradient. Some of the deuterons encounter a CaO crystallite during the diffusion process. After hopping into one of the potential wells provided by the CaO-metal interface volume, it tunnels through a low potential barrier to an adjacent potential well within the interface's 2-dimensionally ordered periodic subsystem. Because of the low potential barrier between potential wells of the 2-dimensional symmetry interface sub-region, and because of the non-self-trapping character<sup>4</sup> of the trapping sites, this tunneling splits the wave function into two coherent parts, as described in Merzbacher's treatment of a particle in a "double-oscillator" potential.<sup>6</sup> The Merzbacher solution of the 2-potential well problem describes a single particle which is split between 2-potential wells. The split-particle wave function has the same wave function phase-coherency that characterizes a single particle bound inside a single potential well. Thus, the transition between single well geometry and double well geometry preserves wave-function phase order. Applied to the Iwamura geometry, it transiently creates a double-maximum coherent deuteron configuration. Further tunnelings further coherently delocalize the wave function, completing a transition from a double-maximum wave function into a coherently partitioned  $N_{\text{well}}$ -maximum Bloch form.

## 7. Cautions

It is probably a mistake to assume that cold fusion processes are always associated with 2-dimensional symmetry Bloch deuterons. The heat release associated with Iwamura transmutations is in the low mW range, whereas a number of F-P related studies have resulted in heat release in the multiple Watt range. The Iwamura Cs studies do not include calorimetric measurement of heat; hence it is unknown to what extent additional cold fusion heat release occurs concurrently with transmutation heat generation. For F-P studies involving bulk Pd, it is the consensus view that measurable excess heat is normally produced in  $PdD_x$  with  $x > 0.85$ . At  $x > 0.7$ , it becomes thermodynamically possible to begin to have some small fraction of the interstitial deuteron population occupying 3-dimensional-symmetry bulk-Pd tetrahedral sites.<sup>7</sup> Some modelers suggest that tetrahedral site occupation by a fraction of the interstitial deuterons present in the bulk is required for F-P heat generation.

It is important to consider the work of Arata and Zhang (A-Z). A-Z have studied excess heat generation occurring in deuterided Pd in the form of fine

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Recently A-Z have ex Pd-Zr alloys.<sup>10</sup> These pow the absorbed gas is absorl ratio approaches 3, i.e., th in English, Arata has sugl powders have fcc structur than one deuteron. Thes the large area/volume rat to that of bulk metal. I two tetrahedral sites and occupied octahedral site can be transiently occup incremental occupation o occupied. In this view, tl in each unit cell forms  $\epsilon$  atom cluster. It would co potential wells. In such  $\epsilon$  could occur in the same n CaO-Pd interface volum coherent Bloch form.

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powders, such as Pd-black.<sup>8</sup> This powder has a nm-scale structure, as evidenced by a broadening of its X-ray Bragg reflection peaks. A-Z have suggested that the powder may include domains having atom cluster order, as described by Fujita.<sup>9</sup> They have also suggested that high surface mobility is involved, and have used the term "spillover deuterium" to describe this property. "Spillover deuterium" is a term used in catalyst literature to describe anomalously high hydrogen activity.

Recently A-Z have explored use of powders produced by oxidizing amorphous Pd-Zr alloys.<sup>10</sup> These powders are very effective absorbers of deuterium gas. If all the absorbed gas is absorbed in the Pd fraction, which is not oxidized, the D/Pd ratio approaches 3, i.e., the  $\text{PdD}_x \rightarrow \text{PdD}_3$ . In a recent paper<sup>11</sup> not yet published in English, Arata has suggested that the structures produced by deuteriding these powders have fcc structures in which the octahedral sites are occupied by more than one deuteron. These expanded structures are presumably made possible by the large area/volume ratio of the powder, which reduces expansion stress relative to that of bulk metal. In his view, the potential well volumes surrounding the two tetrahedral sites and the incrementally available volume associated with a fully occupied octahedral site are about the same. He says that the tetrahedral sites can be transiently occupied. By the same argument the available volume for an incremental occupation of a fully occupied octahedral site can also be transiently occupied. In this view, the presence of these three transiently occupyable volumes in each unit cell forms an interconnecting network that runs through the metal atom cluster. It would constitute a 3-dimensional symmetry network of low barrier potential wells. In such a network the "Merzbacher" coherent partitioning process could occur in the same manner as occurs within Iwamura's 2-dimensional symmetry CaO-Pd interface volume. Diffusing deuteron "visitors" would be converted into coherent Bloch form.

It is also important to note that an ordering of wave function phases should be able to occur without sequential tunneling in sufficiently low temperature Pd which has sufficient lattice array order. In the low temperature limit  $T \rightarrow 0$ , spontaneous ordering should occur in fully loaded palladium PdD. In PdD<sub>1+ $\delta$</sub> , the  $\delta$ -deuteron fraction should exist in a coherently partitioned Bloch form so as to avoid the free energy cost of breaking lattice symmetry.<sup>12</sup> The F-P cold fusion could then take place in a mode that does not break lattice symmetry in a Mossbauer-type momentum-preserving process. Reaction energy goes into the recoil energy of the coherently partitioned Bloch product  ${}^4\text{He}_{\text{Bloch}}^{2+}$  and the recoiling crystallite. This type of fusion does not require the irreversibility step associated with electron scattering. For example, a recoiling host occurs during the Mossbauer energy release from an iron-containing lattice in a process that balances the momentum of a released gamma ray. The de-excited Fe atom recoils as part of the recoiling lattice crystallite. The spontaneous ordering would be an example of the ordering that occurs in many-body lattice systems.<sup>13</sup> However, the author's present view is that it is unlikely that spontaneous ordering is occurring in the above-room-temperature environment that characterizes the Iwamura process.

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